RESEARCH AND TECHNOLOGIC WORK ON COAL AND RELATED INVESTIGATIONS, 1957

BY BUREAU OF MINES STAFF

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RESEARCH AND TECHNOLOGIC WORK ON COAL AND RELATED INVESTIGATIONS, 1957[1]

by

Bureau of Mines Staff

INTRODUCTION AND SUMMARY

This report is the 22d in a series summarizing Bureau of Mines research and technologic work on coal and related investigations.[2 3] A summary of work published during 1957 and of work in progress is presented. Details regarding the subjects treated in the publications may be found by referring to articles cited in the footnotes.

The objective of the coal work of the Bureau of Mines is to obtain information that will lead to improved production and utilization of coal, thus conserving a most important national asset.[4 5] Though studies of mining methods and exploration of various coalfields may contribute directly to improved production, no less so does the work in health and safety. The many problems relating to chemical and physical properties of coal reflect the need for an understanding of these properties for the most effective use of coal in its various fields of application and for the development of new methods of processing coal. Studies of existing and proposed processes of combustion, carbonization, gasification and hydrogenation of coal are necessary for increased efficiency in its use. Engineering service provided by the Bureau is of direct value in the conservation program in preventing waste. Stations where the main coal activities of the Bureau are carried out are shown in the following list:

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[1] Work on manuscript completed October 1958.

[2] Staff, Bureau of Mines, Report of Research and Technologic Work on Coal and Related Investigations, 1956: Bureau of Mines Inf. Circ. 7904, 1959, 112 pp.

[3] Carman, E. P., Geer, M. R., and Riley, H. L., Report of Bureau of Mines Research and Technologic Work on Coal and Related Investigations, 1955: Bureau of Mines Inf. Circ. 7794, 1957, 103 pp.

[4] Cohn, E. M., Fuels: The Encyclopedia of Chemistry (George L. Clark and Gessner G. Hawley, eds.), Reinhold Pub. Corp., New York, N. Y., 1957, pp. 426-427.

[5] Cohn, E. M., United States Bureau of Mines: The Encyclopedia of Chemistry (George L. Clark and Gessner G. Hawley, eds.), Reinhold Pub. Corp., New York, N. Y., 1957, pp. 961-962.

Station	Location	Main coal activities
Headquarters	Washington, D. C.	Programing and budgeting; foreign activities; Government fuel-engineering service.
Anthracite Exp. Sta.	Schuylkill Haven, Pa.	Anthracite investigations.
Region I		
Alaska Mining Exp. Sta.	Juneau, Alaska	Exploration.
Anchorage Exp. Sta.	Anchorage, Alaska	Sampling and analysis.
Northwest Exp. Sta.	Seattle, Wash.	Preparation.
Region III		
Denver Exp. Sta.	Denver, Colo.	Western coals investigations.
Lignite Exp. Sta.	Grand Forks, N. Dak.	Lignite investigations
Region V		
Central Exp. Sta.	Pittsburgh and Bruceton, Pa.	Health and safety investigations (District B); mining, preparation, combustion, carbonization, hydrogenation, petrography and constitution, sampling and analysis of bituminous coal.
Appalachian Exp. Sta.	Morgantown, W. Va.	Gasification, low-temperature tar.
Eastern Exp. Sta.	College Park, Md.	Boiler-water research and service; stream and air pollution.
Southern Exp. Sta.	Tuscaloosa, Ala.	Preparation and carbonization.
Gorgas Exp. Sta.	Gorgas, Ala.	Underground gasification.

The Bureau receives many requests yearly from Federal agencies for coal analyses to aid in evaluating bids for purchased coal. A summary of analyses of tipple and delivered samples was published in 1957, the seventh in this series.

Two international systems for classifying coals - one for higher rank and the other for lower rank coals - have been recommended by the Coal Committee of the Economic Commission for Europe. The Bureau cooperated in developing both systems. American coals can be assigned to at least 26 of the 59 code numbers of the international system for the higher rank coals.

Methods for analyzing coal and derived products are under continuous review, and new methods are being developed. The ash contents of certain coals, containing calcite and pyrite, were found to depend on the rate of heating during the ashing procedure. In a study of the chlorine contents of coals from 15 States and Alaska, coals from the Western States and Alaska were found to contain no chlorine or very small quantities, whereas those from the Eastern States contained amounts that varied with and within the bed. New and more rapid methods have been developed for analyzing coal ash and related materials. Methods of applying spectrophotometry to various analyses are being studied.

Knowledge of the molecular structure of coal is being obtained by studies of its optical properties in the visible, infrared, and ultraviolet regions, by X-radiation, and by nuclear magnetic resonance measurements. Coals have been separated into relatively pure petrographic components; these components when heated have shown different chemical analyses, X-ray patterns, and behavior.

Researchers developed two new methods for hydrogenating coal at low temperatures - one by using lithium in ethylenediamine and the other by an equimolar mixture of carbon monoxide and hydrogen in the presence of dicobalt octacarbonyl. The results obtained suggest that further studies of the products from these reactions will help in elucidating the chemical structures present in coal. Studies of microbial degradation of coal have been continued to develop another low-temperature process to yield products that have not been altered by the application of heat; the bacteriostatic and fungistatic materials found in coal in the course of these studies have been investigated further. To aid in solving problems of coal structure, the properties of naturally occurring humic acids from large deposits of oxidized lignite from North Dakota are being compared with those of acids made in the laboratory at higher temperatures.

Churn and core-drilling operations were continued in Alaska throughout the 1957 field season in the western part of the Wishbone Hill district of the Matanuska area to obtain data on the location and character of the coal measures. Reconnaissance was made of the less accessible parts of the Beluga coalfield, near Anchorage.

A principal objective of the Bureau of Mines is to develop improved and safer mining methods. Studies were continued of the use of a Bureau-designed pneumatic coal planer in longwall mining of anthracite. The scraper-shaker loader - also designed by the Bureau - removed blasted material in one-fourth the time with one-fourth the number of men, thus achieving a sixteenfold improvement over hand-loading methods. Cooperation in a joint Federal-State program for the control of water in anthracite mines has resulted in the approval of 14 additional projects in 1957.

Operations at five bituminous coal mines have shown that longwall mining with a German coal planer can be successfully carried out in thin beds of coal with increased recovery. A study of methods and equipment for underground development in 14 bituminous coal mines was made, and the results were published.

Data obtained from the operations at eight mines showed that, although coal can be produced cheaply by auger mining, the recovery ranges from only 20 to 50 percent. Fieldwork is in progress to compare roof bolting with timbering for mine roof support. Studies have been made in 12 mines in which various combinations of methods of face haulage and equipment can be used in conjunction with continuous-mining machines.

The percentage recovery of coal by various mining methods determines the extent of reserves of useful product; this is a subject of continuing study. Recent calculations from data obtained from five mines operated in the Upper Freeport and from two mines in the Pittsburgh bed showed that the recovery ranged from 50 to 60 percent where the pillars were not recovered and from 80 to 88 percent where they were recovered.

Significant data have been collected in mines operating in the Pittsburgh bed where test zones of varied roof-bolting patterns have been established. Approximately 80 percent of all roof fall fatalities occur beyond the point where the bolts were installed. Roof bonding is also under investigation as a means of increasing roof stability. The continuing study of new developments for improving roof control is a necessary consequence of the fact that roof falls are the major cause of fatalities in coal mining. An analysis of the conditions associated with 1956 fatalities from roof falls was published during the period covered by this report. A similar analysis was published on haulage fatalities. A summary and analysis of reports from coal-producing companies and State mine officials on injuries and related employment data in the coal-mining industry for the years 1952 through 1955 show a progressive and gradual improvement over the preceding years.

Recommended safety standards for shaft sinking have been compiled and published because so many unfortunate occurrences have resulted from this operation in the past few years. In connection with its program of improving mine safety, the Bureau has contributed to the development of a safer trailing cable (the trailing cable is the most serious mine-fire hazard), mine lighting, communication equipment for use in emergencies following mine fires and explosions, and auxiliary and supplemental mine rescue equipment. Investigations were continued on major ventilating problems confronting the industry and on dust control in mines.

Among the many technical problems associated with coal after it is mined and before it is used storage and preparation are receiving special attention. Lignite withstands storage less satisfactorily than higher rank coals. A unique opportunity of studying the storage of lignite was provided when 2 million tons of lignite was mined and stored in piles when Garrison Dam was being constructed in North Dakota. Continuous observations have been made; during the year it has been shown that the heating value of the coal, on a moisture- and ash-free basis, has decreased less than 1-1/4 percent from August 1953 to August 1957 and that the change in size consist has been slight. These facts may be attributed to the high compaction during piling, with bulk densities approaching 70 lb. per cu. ft.

Lignite is a coal that contains a high percentage of moisture when mined. Unloading cars shipped in subfreezing weather is often difficult due to agglomeration of the coal by freezing in the cars. To evaluate the freezeproofing qualities of various reagents, a reproducible procedure has now been developed.

The tough, resilient qualities of raw lignite make it difficult to pulverize. Technologists are investigating various methods of pulverization.

Present methods of cleaning fine coal are less effective and more costly than treatment of the coarser sizes. To minimize the waste of fine coal now lost through ineffective cleaning, studies are in progress on flotation, the dense-medium cyclone, and the feldspar jig. The first installation of a modern feldspar jig in this country was made at a mine in Washington State, where it has been subjected to a number of performance tests by the Bureau.

The suspended fine coal in the water leaving a coal-washing plant is difficult to remove to the extent required to meet stream-pollution requirements. Federal researchers are studying methods for flocculating this coal.

For some years the Bureau has studied the preparation characteristics of coals on a county basis. Three additional reports were issued in 1957, bringing the total completed to 21 counties. These reports have special value in delimiting the reserves of coal that meet present standards required for use in producing metallurgical coke.

A laboratory for studying special problems in preparing anthracite was established; coal fractions containing as little as 0.75 percent ash have been prepared. Combustion of coal is its principal use. Many problems arise from ash associated with the coal, from water used to transfer the energy to a turbine that converts it into power, and from effluents from the combustion chamber that constitute potential air pollution. Ash deposits form on surfaces of boiler tubes, where they reduce heat flow and hinder gas flow through the furnace. The mechanism involved in formation of these deposits is being studied both by analyses of data obtained in large steam-boiler furnaces and in laboratory equipment.

During the period covered by this report, tests have been made on the performance of four small, industrial anthracite stokers to supplement information available on combustion of anthracite in domestic stokers and in large powerplants.

Return of steam condensate for boiler feedwater may result in large maintenance costs owing to corrosion of pipes and valves. A Bureau condensate-corrosion tester is widely used to determine the need for corrective measures and to evaluate the effectiveness of filming treatment. Investigations included filming treatments using octadecylamine and its acetate derivative. Engineering service was given to Federal agencies on water problems.

Air pollution resulting from combustion of coal and refuse and from automobile exhaust has been studied with laboratory equipment. The principal contaminant from the combustion of coal is sulfur dioxide; thus, researchers studied several processes for its removal. In the combustion of refuse an added problem is the removal of obnoxious odors. Work is in progress on the design and operation of incinerators capable of producing completely innocuous discharges from refuse and radioactive wastes. Equipment has been assembled to study the prevention of air pollution by catalytic oxidation of exhaust gases from an automobile engine, a complicating factor is poisoning of the catalyst by lead deposits from leaded gasoline.

In carbonization studies different laboratories of the Bureau use equipment designed to give the most information on the particular problem studied. The scale of operation ranges from a few grams to several hundred pounds per test. Small-scale studies of the mechanism of coke and tar formation are being made in the temperature range of 300° to 550° C. This same problem is being studied on a larger scale in a fluidized bed. Measurements of expansion of coal are also made on two scales of operation - one test on 71 grams of coal and the other on 18 kilograms; good correlation has been found between the two tests.

Further work has confirmed previous findings that pretreatment of coals at temperatures below their softening points improves the physical properties of the resulting cokes. Carbonization of the hot pretreated coal gives results superior to those obtained when the pretreated coal is cooled before carbonization.

A program was initiated to study the effect of cleaning on the coking properties of coal, and special consideration will be given to possible changes in petrographic composition of raw and cleaned coal.

In a study of two coals that differed greatly in fluidity abradabilities of their cokes were found to be almost identical when their fluidities were reduced to about five dial divisions per minute by addition of an inert.

The study of improvement of the quality of coke made from western high-volatile bituminous coals has been continued in a 14-inch slot-type oven. The BM-AGA survey of carbonizing properties of American coals was extended to include 22 additional coals from West Virginia and 4 coals from Pennsylvania. Publications released during the year included surveys on coals from one county in West Virginia and two counties in Pennsylvania.

For many years the use of anthrafines in the coal charge to an oven for producing metallurgical coke has been of interest, not only for improving coke quality but also for marketing the finer sizes of anthracite. Work on this problem was extended to several blends of high- and low-volatile coals with a constant amount of anthrafines from a single source. Several blends yielded cokes having essentially the same physical properties.

Studies were continued in using anthracite in blast-furnace and cupola operation, with or without thermal stabilization by calcination. A pilot-scale vertical-shaft calciner having a capacity of 10 to about 500 pounds per hour was constructed.

The effects of calcining depend on the quality of the anthracite feed. The products were tested in a cupola. Because the product of calcination does not meet the currently accepted strength specifications determined by ASTM methods, work is in progress to study the production of briquets from anthracite, bituminous coal, and coal-tar pitch that will meet these specifications after calcination.

In recent years interest has revived in low-temperature carbonization of coal. Such carbonization produces char, useful as a fuel in generating power, and large yields of tar, a source of chemicals and pitch for electrode manufacture. To provide information on the yields of low-temperature products of American coals, a bulletin was published that gives the results of Fischer-Schrader low-temperature assays at 500° C. of about 400 coals from nearly every coal-producing State, Alaska, and British Columbia. A different small-scale carbonization assay test, originally reported in 1953, is now routinely applied. Chars have been prepared from Wyoming noncoking or slightly coking coals by using both bench-scale and pilot-plant techniques to determine whether they might be suitable as blending material for metallurgical coke manufacture, used in phosphate ore reduction, converted to activated carbons, or employed as boiler fuel. Further low-temperature carbonization experiments on a dried Texas lignite were carried out in an 8-inch fluidized-bed retort to determine the effects of process variables.

The Bureau continued its program of technical assistance to the Indian Government in connection with utilization of lignite from Neyveli, South Arcot. It was shown that this lignite could be dried satisfactorily in a fluidized bed and briquetted at high pressures without a binder. An integrated pilot plant was designed and procured for installation at the mine site at Neyveli.

Low-temperature carbonization depends for its economic success on producing tar that has a value greater than its fuel value, either through direct fractionation to materials of interest to the chemical markets or through upgrading by secondary processing. Low-temperature tars from lignite and subbituminous and bituminous coal are being intensively studied. Detailed studies of the phenols and bases present in the tars have been made, and similar work was begun on the neutral fraction. Novel techniques have been developed, and certain generalities regarding the composition of low-temperature tar are beginning to emerge. The work to date on upgrading the tar has been concentrated on physical separation methods, although work on chemical conversion processes is included in the program.

To supplement the future supply of natural gas and petroleum, coal may have to be gasified to provide pipeline gas and liquid fuels. Because the rate of gasification of coal is limited by the rate of reaction of steam with carbon, this reaction has been studied under many conditions for several years. Gasification of coal yields hydrogen and oxides of carbon in varying ratios, depending on the conditions of operation; if pure hydrogen is the product sought, the primary gas is subjected to a second reaction with steam to "trade" carbon monoxide for hydrogen. An alternative process has been investigated - the steam-iron technique in which iron oxide is reduced by coal-derived gases, and iron is then reacted with steam to regenerate iron oxide and yield hydrogen. Chemists are investigating reaction rates in this process to find optimum conditions of operation.

Lower rank coals offer two principal advantages over higher rank coals as raw materials for gasification: They do not agglomerate and are more reactive. Results have been published on gasifying lignite in a small-scale, externally-internally heated annular-retort gasifier. More recently, activity has been concentrated on developing a process for gasifying lump fuel in a fixed-bed gasifier with oxygen and steam at elevated pressure. Because hydrogen will be produced under pressure in this process, studies have been made of the agglomerating tendencies of lignite at pressures of 600 to 1,000 p.s.i.g. of hydrogen.

For several years, gasification of pulverized coal has been studied in a fluid-bed pilot plant. Changes in design have been introduced periodically to improve operations. One of the major problems, feeding pulverized coal to a pressurized system, has been investigated by several approaches.

Gasification consumes heat; hence heat must be supplied to the reactor. This heat can be transferred through a metallic coil in which coal is reacted with steam. An attractive source of heat is nuclear energy. Heat from nuclear reactors might be used in several ways, and technologists are studying the problems associated with them. Emphasis has been placed on materials of construction to be used in these high-temperature reactions and on assorted problems of transferring heat.

Anthracite was gasified in a Lurgi gasifier at the Dorsten Works of Steinkohlengas A.G. in Germany. Further development is required before a coal-to-pipeline gas process can be based on anthracite as the fuel.

Studies of underground gasification of coal were continued, and additional experiments on hydraulic fracturing and electrolinking were carried out.

Gas made from coal contains gaseous impurities and dust, both of which must be removed before the gas can be processed further into end products. Hot solutions of potassium carbonate are a good purification medium. Studies are in progress to develop methods for generating hot dust-laden gas, for sampling, and for removing dust at elevated temperatures and pressures.

Studies of the hydrogenation of coal under pressure at elevated temperatures to produce liquid fuels have had an important position in the Bureau's coal program for many years. In this work use has been made both of autoclaves and of pilot-plant equipment. The principal use of autoclaves in the period covered by this report has been in a study of the mechanism of the iron-catalyzed hydrogenation of coal. Work in the pilot plant has been directed toward output of distillable oils for possible use in turbines and jets rather than production of gasoline for use in internal combustion engines. Lower pressures, which should reduce the cost of plant significantly, have been used.

Equipment was also designed and used to study the hydrogenation of coal at very short reaction times and at about 800° C. A continuous unit for hydrogenating coal under these conditions has been partly assembled.

A significant advance has been made in methods for characterizing the products from coal hydrogenation in the development of a method of mass spectrometric analysis using a low ionizing voltage.

An alternative to coal hydrogenation for producing synthetic liquid fuels involves intermediate gasification to a synthesis gas containing hydrogen and carbon monoxide and subsequent conversion to liquid products by catalytic reactions. The chemistry of metal carbonyls is being explored to develop a better understanding of the mechanism of the Fischer-Tropsch synthesis. Studies of the Fischer-Tropsch catalysts have received major attention. Characterization of the products obtained under various conditions has continued and has contributed significantly to an understanding of the nature of the reactions involved. Pilot-plant development of the hot-gas recycle process has shown that this system both simplifies construction and decreases the cost of reactors.

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Allegheny Pittsburgh Coal Co.
American Colloid Co.
American Electro Metal Div. of Firth Sterling, Inc.
American Society of Mechanical Engineers
Ampco Metal, Inc.
Atlantic Refining Co.
Borolite Corporation
Boslego Coal Co.
Carbide and Carbon Chemicals Co., Div. of Union Carbide Corp.
Carbone Corp.
Carborundum Co.
Chromalloy Corp.
Clark Bros., One of the Dresser Industries
Clinchfield Coal Corp.
Colorado Fuel and Iron Corp.
Colorado School of Mines Research Foundation, Inc.
Columbia-Geneva Steel Div. U. S. Steel Corp.
Comision de Fomento Industrial, Cali, Colombia
Comision de Fomento Minero, Mexico, D. F.
Consolidation Coal Co. of W. Va.
Crucible Steel Co. of America
Dakota Briquets & Tar Products, Inc.
Djawatan Pertambangan (Indonesian Bureau of Mines), Djakarta, Indonesia
Duquesne Light Co.
Eastern Gas & Fuel Associates
Electro Metallurgical Co., (A Div. of Union Carbide Corp.)
El Paso Natural Gas Products
Emerald Coal and Coke Co.
Fansteel Metallurgical Corp.
Ford Motor Co.
General Electric Co.
Glen Alden Coal Co.
Halliburton Oil Well Cementing Co.
Hanna, M. A., Co.
Hudson Coal Co.
Ingersoll-Rand Co.
International Cooperation Agency (Neyveli, India, Project)
Island Creek Coal Co.
Joanne Coal Co.
Johnstown Coal & Coke Co.
Joy Manufacturing Co.
Kaiser Steel Corp.
Lava Crucible Refractories Co.
Lehigh Valley Coal Sales Co.
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Mine Safety Appliances Co.
Minnesota Mining and Manufacturing Co.
Minnkota Power Cooperative, Inc.
National Carbon Co., Div. of Union Carbide Corp.
Neyveli Lignite Corp. Central Govt. of India, and Madras State Govt.

Northern States Power Co.
Norton Co.
The Pacific Coast Co.
Pan-American Petroleum & Transport Co.
Peabody Coal Co.
Petrocarb Equipment, Inc.
Pittsburgh Consolidation Coal Co.
Powhatan Mining Co.
Reading Anthracite Co.
Republic Steel Corp.
Rich Hill Coal Mining Corp.
Riley Stoker Corp.
Rochester and Pittsburgh Coal Co.
Royal Afghan Ministry of Mines, Kabul, Afghanistan
Saginaw Dock & Terminal Co.
Standard Oil Co. of N. J.
Steinkohlengas A. G.
Susquehanna Collieries Co.
Texas Power & Light Co.
Thompson Products, Inc.
Trotter Coal Co.
Traux-Traer Coal Sales Co.
Union Carbide Corp.
University of Alabama
University of North Dakota
University of Washington
U.S. Army Corps of Engineers
U.S. Atomic Energy Commission
U.S. Geological Survey
U.S. Pipe & Foundry Co.
U.S. Public Health Service
Warner Collieries Co.
West Virginia University
Youghiogheny & Ohio Coal Co.

ORIGIN AND PROPERTIES OF COAL

Inspection, Sampling, and Analysis

The Bureau of Mines maintains an inspection, sampling, and analysis service for all Federal agencies, including the Bureau itself. It makes available analyses of tipple and delivered samples accumulated in the course of this work to the public, and these analyses are widely used in the purchase of industrial coal. In 1957 the seventh[6] of a series of annual publications was released, covering analyses of tipple and delivered samples of coal collected throughout the United States and Alaska. These analyses are arranged alphabetically with respect to States, counties, towns, and mines and are listed as follows: Proximate analysis, ultimate analysis, calorific value, ash-softening temperature, agglomerating index, free-swelling index, and Hardgrove grindability index. Engineers of the Bureau of Mines took tipple samples at the mine tipple or breaker as the coal was loaded into railroad cars and trucks. Representatives of Government departments, under directions supplied by the Bureau of Mines, systematically collected samples throughout all deliveries.

Statistics placed the coal requirements of Federal agencies, except AEC and TVA, for 1957 at approximately 6 million tons. All agencies but the Post Office Department purchased their coal on a guaranteed-analysis basis, including penalties for delivery of sub-standard coal. Requests from Federal agencies for analyses to be used in evaluating coal bids continued at about the same rate as in the preceding year because of the large number of bids and strict adherence to specification requirements regarding elimination of bids from mines on which there were no Bureau of Mines analysis records. Contracting for coal for the fiscal year 1958 was started in January and continued for the balance of the fiscal year. Recommendations were made for awarding contracts for some agencies.

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[6] Aresco, S. J., Haller, C. P., and Abernethy, R. F., Analyses of Tipple and Delivered Samples of Coal (Collected During the Fiscal Year 1956): Bureau of Mines Rept. of Investigations 5332, 1957, 67 pp.

Proximate or ultimate, sulfur and calorific value analyses were reported on 10,922 samples, a decrease of 2.5 percent over the calendar year 1956; ash-softening temperatures on 2,318 samples; free-swelling index on 368 samples and Hardgrove grindability on 140 samples from purchases of coal and tipple and breaker inspections collected in connection with Government coal purchases. Analyses were reported on 783 tipple samples collected at 311 mines in 16 States and on 373 samples of export coal, approximately 579,300 gross tons, being shipped overseas by the ICA. At the request of the General Services Administration, four cargoes were sampled at Eastern and Gulf ports. Twenty-five carbonization samples and 29 preparation samples were collected at mines in Pennsylvania and West Virginia.

At the request of the Department of the Navy, size-consist samples were collected at the New York Naval Shipyard, Brooklyn, N. Y., and personnel at the installation was instructed in proper coal-sampling procedures. Size-consist samples of run-of-mine coal for Rosin-Rammler plots were collected at mines in Iowa, Maryland, Oklahoma, and Pennsylvania. Under the working agreement with AEC, 491 samples of delivered coal were collected at the Savannah River Operations Plant, Aiken, S. C., and 124 samples were collected at the Dana Plant, Ind. Sampling at the Dana plant ended in May when the plant was put on a standby basis.

Meetings were held with representatives of the Air Force, Army, and Navy to discuss coal-sampling problems. Engineers conducted a series of tests on the use of a multiple riffle for use at AEC's Savannah River Plant, Aiken, S. C. No conclusion has been arrived at on the use of this type riffle, pending the results of samples being tested in the laboratory. To fill a pressing need, the agency revised directions for coal sampling.[7] The Bureau has retained most of the original material pertaining to the hand and mechanical methods of preparing samples. It has added instructions for obtaining "top of car" samples, minimum number and weights of increments to be collected for various sizes of coal sampled and for filling out its Form 6-220.

The Anchorage Laboratory was authorized, on February 25, 1957, to make, certify, and report all military coal-delivery samples originating in Alaska. A total of 841 samples from several sources were prepared. Six tipple samples were collected during the year from five of the six mines now filling military contracts. Periodic visits were made to the Army or Air Force bases to inspect sampling facilities, assist with coal-sampling procedures and problems, and train samplers.

During 1957 nearly 32,000 samples were analyzed involving over 275,000 determinations. The samples included coals purchased by the Government on a guaranteed-analysis basis, tipple samples, and samples submitted by Bureau of

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[7] Snyder, N. H. (revised by S. J. Aresco), Coal Sampling: Bureau of Mines Handbook, 1957, 16 pp.

Northern States Power Co.
Norton Co.
The Pacific Coast Co.
Pan-American Petroleum & Transport Co.
Peabody Coal Co.
Petrocarb Equipment, Inc.
Pittsburgh Consolidation Coal Co.
Powhatan Mining Co.
Reading Anthracite Co.
Republic Steel Corp.
Rich Hill Coal Mining Corp.
Riley Stoker Corp.
Rochester and Pittsburgh Coal Co.
Royal Afghan Ministry of Mines, Kabul, Afghanistan
Saginaw Dock & Terminal Co.
Standard Oil Co. of N. J.
Steinkohlengas A. G.
Susquehanna Collieries Co.
Texas Power & Light Co.
Thompson Products, Inc.
Trotter Coal Co.
Traux-Traer Coal Sales Co.
Union Carbide Corp.
University of Alabama
University of North Dakota
University of Washington
U.S. Army Corps of Engineers
U.S. Atomic Energy Commission
U.S. Geological Survey
U.S. Pipe & Foundry Co.
U.S. Public Health Service
Warner Collieries Co.
West Virginia University
Youghiogheny & Ohio Coal Co.

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[7] Snyder, N. H. (revised by S. J. Aresco), Coal Sampling: Bureau of Mines Handbook, 1957, 16 pp.

Mines and other Government organizations in connection with research and exploration projects on coal. The largest number of samples (18,618) was submitted by the Federal Coal Mine Inspectors from 3,440 inspections in 21 States and Alaska; the incombustible content was determined for all the coal-mine dust samples to check conformity with the provisions of Public Law 552, Federal Coal Mine Safety Act.

International Classification of Coal

In 1949 the Coal Committee of the Economic Commission for Europe established a Classification Working Party to undertake development of an international system for classifying coal. Since that time experts from nearly every European country and the United States have actively participated in the work. The Bureau of Mines has cooperated both by attendance of delegates at meetings and by laboratory investigative work.

A system for classifying the high-rank coals was approved in 1956, with the recommendation that the system be given practical application. In the study of the application of the system to American coals, analyses and small-scale caking and coking tests have been made of nearly 100 samples of representative coals of low-, medium-, and high-volatile bituminous rank. It was found that American coals can be assigned to at least 26 of the 59 code numbers of the international system. In 1957, a system for classifying lower rank coals (mainly brown coals and lignites) was formulated with the recommendation to the various Governments that the system be tried for 1 year.

New Analytical Procedures

Many methods for analyzing coal and derived products have been developed by the Bureau and accepted as standards. Not only are these methods under continuous review but new methods are being investigated.

As part of a biennial series that appears in Analytical Chemistry a review was prepared of new developments in methods for analyzing and testing solid and gaseous fuels covering the period from July 1954 to October 1956.[8]

A series of coals was ashed by both American Society for Testing Materials (ASTM) and proposed International Standards Organization (ISO) procedures. This work proved that the rate of heating in the proposed ISO procedure was too rapid for coals containing calcite and pyrite. The faster rate of heating fixed more of the sulfur in the ash. There was close agreement in ash values by the two methods after correction for this sulfur. This work was presented to ISO through the normal channels of ASTM and the American Standards Association.

Forms of sulfur were determined in five coals pulverized to 60- and 200-mesh to evaluate the effect of particle size on the extractions. Results of the tests show that there is no advantage in pulverizing to 200-mesh. Another reason for reviewing this test is that certain types of low-rank coals give erroneous results.

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[8] Ode, W. H., Solid and Gaseous Fuels, Industrial Applications of Analysis, Control, and Instrumentation: Anal. Chem., vol. 29, No. 4, April 1957, pp. 657-669.

There is no standard method of testing for sulfur in coal ash. A test is needed for making certain ash corrections and for estimating the oxygen content of coals. In developing a standard procedure for determining sulfur in coal ash several analytical applications were employed. Preliminary work showed that hydrogen sulfide was lost when the ash was treated with hydrochloric acid. Later work indicated that ash obtained by the standard method contained insignificant amounts of sulfides soluble in hydrochloric acid.

Several analytical methods for determining chlorine in coal and organic materials were examined for speed in operation and for accuracy. The method adopted consists of burning the sample in an oxygen bomb; the chlorine is measured by an electrical titrimeter, using silver nitrate as a titrant. Two hundred and thirteen analyses for samples from 15 States and Alaska have now been made. Coals from Colorado, Montana, New Mexico, North Dakota, Utah, Washington, Wyoming, and Alaska contain no chlorine or very small quantities. If analyses from other localities in these States show such small quantities of chlorine, the testing of samples from these States will be discontinued. Chlorine in the bituminous coals of the interior and eastern provinces varies from bed to bed in the same locality and for the same bed in different localities.

Modern physicochemical techniques have been applied in studies of development of rapid, routine methods for analyzing coal ash and related materials, such as slags, flue dusts, and external boiler-tube deposits. Classical methods of quantitative analysis, used for many years, are relatively slow and not always adaptable to rapid analysis of large numbers of samples. The investigation has shown that routine analyses of ash and slag can be obtained quickly and economically by colorimetric methods for silica, by flame-photometry for sodium and potassium, or by complexometric titration for calcium and magnesium. These methods gave results that are accurate enough for most applications.

Empirical formulas are now used to convert the ash content of coal to mineral matter content, which is needed for calculation of coal analyses to a dry, mineral matter-free basis. Two methods for direct determination of mineral matter have been investigated: (1) A method developed in Germany to remove the bulk of the mineral matter by treatment with acids and corrections for residual ash, for pyrite not removed, and for chlorine taken up by the coal was found to be satisfactory for all ranks of American coals tested, provided the method is modified slightly for coals containing calcite; (2) solvent extraction of coal to obtain the mineral matter in a form suitable for estimation and identification of the various constituents has been investigated but has not yet been fully developed.

Correlations of ultraviolet-visible spectra and molecular structure have proved useful in analytical work and in understanding the relationship of structures of various aromatic compounds.[9 10]

Quantitative applications of infrared and ultraviolet vapor spectrophotometry have been developed.[11 12] Accurate sample introduction by the micropipet - sintered-disk technique circumvents difficult measurement of vapor pressures. The method is applicable to compounds of low volatility because low vapor pressures are usable.

A new extruding device permits preparation of small cylindrical specimens suitable for X-ray powder-diffraction analysis without the use of binder.[13] This device is particularly suitable and useful for studying organic materials, and preparation of specimens weighing as little as 2 or 3 mg. is feasible. These techniques in modified form serve for preparing even smaller specimens, perhaps of 0.1 or possibly 0.01 mg.

Structure and Chemistry of Coal

Indexes of Refraction and Absorption

Previous work has shown that these optical properties differ for the various components of single coals, varying with the rank of the coal, and may be useful in the selection of coals for carbonization, hydrogenation, and combustion. Maximum and minimum indexes of refraction and absorption of the vitrinite components of coals of different rank were determined from microscopic reflectivity measurements in different media. Similar determinations were also made on single crystals of graphite, because most theories of coal structure assume that it is the end product of the coalification series. As far as known, this is the first time that the optical constants of graphite have been determined in various directions relative to the layer planes. For the coal-graphite series, a relationship was found between both the reflectivity and the index of absorption and the hydrogen-carbon ratio, a measure of coal rank.

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[9] Friedel, R. A., Correlations in Ultraviolet Spectrophotometry: Applied Spectroscopy, vol. 11, No. 1, November 1957, pp. 13-24.

[10] Friedel, R. A., Book Review of Ultra-Violet and Visible Absorption Spectra: Index for 1930-1954, by Herbert M. Hershenson. Spectrochimica Acta, vol. 10, No. 1, November 1957, p. 131.

[11] Friedel, R. A., Quantitative Ultraviolet Vapor Spectrophotometry: Applied Spectroscopy, vol. 11, No. 2, 1957, pp. 61-64.

[12] Friedel, R. A., and Queiser, J. A., Quantitative Infrared Vapor Spectra: Anal. Chem., vol. 29, No. 9, September 1957, pp. 1362-1366.

[13] Hofer, L. J. E., Damick, A., Headrick, A. F., Fauth, F., Bean, E. H., and Golden, P. L., Extrusion of Cylindircal Specimens for X-ray Powder Diffraction Analysis: Anal. Chem., vol. 29, No. 10, October 1957, pp. 1563-1564.

Spectroscopic Studies

Ultraviolet and visible absorption spectra of various coals have been determined.[14] The absorption is comparatively weak and does not justify the hypothesis that polynuclear condensed aromatics are present in high concentration. Correlations have been found between free-radical content within various ranks of coal and the ultraviolet absorption intensity at various wavelengths. It is proposed that free radicals rather than polynuclear condensed aromatics may be the cause of the color, absorption spectra, reflectivity, and refractivity of coal.

The ultraviolet-visible spectrum of Pittsburgh vitrain and the spectra of two high molecular weight products from coal hydrogenation (asphaltene and heavy oil) have been determined. The decreasing absorption in the visible region from coal to heavy oil correlates directly with the decrease in free-radical content shown by electron paramagnetic resonance measurements. Absorption in the ultraviolet region, however, shows a complete reversal. As small aromatic molecules absorb strongly in this region, the intense absorption of heavy oil and the strong absorption of asphaltene indicate that aromatics are produced, rather than hydrogenated in the coal-hydrogenation process.

Nuclear magnetic resonance measurements have been carried out on asphaltene in an effort to determine the types of protons present. Three absorption peaks indicate the presence of CH_2 and CH_3 groups, some of which are bonded to aromatic or unsaturated systems. Aromatic protons are also indicated; however, no protons are indicated on larger than benzenoid aromatic systems.

Further studies have been made to determine the origin of the strongest infrared band in coal and char spectra. Char prepared from anthracene at 550° C. contained only 0.03 percent oxygen and did not show this strongest band, supporting the previous conclusion that this band is associated with some type of oxygen linkage in coals and chars.

X-ray Diffraction

A critical review of published work on the application of X-rays to studies of the structures of coals was prepared. X-ray scattering studies have made a valuable contribution to knowledge of the complex structure of coal, and refinements in experimental technique and further development of theoretical analysis offer promise of extensive future contributions to the understanding of coal structure.

Because of the complexity of coal structure, much useful information concerning its makeup has been obtained by subjecting coal samples to very mild chemical or thermal reactions to remove less complex fractions that can be more readily identified as to composition and molecular structure. One such process involves heating under vacuum and immediate condensation of the volatilized products. A study was made of the X-ray scattering properties of the vitrinite

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[14] Friedel, R. A., Aromaticity and Colour of Coal: Nature, vol. 179, No. 4572, June 15, 1957, pp. 1237-1238.

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component of a bituminous coal and of the distillate and residues prepared from it by such high vacuum pyrolysis. Comparison of the X-ray scattering intensities indicated that both the distillate and residue contained aromatic layers. Scattering intensities of the distillate and of the residue obtained after 10 percent weight loss were similar and more diffuse than those of the original vitrinite. The present findings can be tentatively explained if the vitrinite is conceived as a three-dimensional polymer of small, partly oriented planar units which depolymerize upon heating; some of the structural entities are set free as vapor, others recombine to form a highly disordered carbon skeleton.

A new method has been developed for determining X-ray absorption coefficients of inhomogeneous materials. It involves comparison of the reflected and transmitted intensities from the same specimen at various scattering angles. The difference in the geometry of the reflection and transmission techniques permits calculation of the absorption coefficient from the observed intensities. Evaluation of the absorption coefficient at different angles overcomes the uncertainties of the conventional method caused by small angle scattering, crystalline mineral impurities, etc., and eliminates the necessity of measuring the intensity of the primary beam.

Petrographic Studies

A report has been published describing the relation between petrographic composition and free-swelling properties of a heterogeneous high-volatile A bituminous coal.[15/] The investigation of this coal showed that banded vitrinite (anthraxylon) is the principal swelling agent, whereas finely divided vitrinite (translucent humic matter) is agglomerating but virtually nonswelling. Micrinite (opaque matter) was essentially inert but in moderate quantities seemed to benefit the residue as a strengthening agent. Fusinite was nonswelling. Microscopic examination of the residues showed that those of banded vitrinite had extremely thin walls, whereas the residues from coal containing a quantity of nonswelling constituents had considerably thicker walls. Such thick-walled residues were smaller and stronger than the banded vitrinite residues. Their strength was due in part to accumulation of the inert ingredients in the cell walls. The differences in properties of these residues suggest the possibility of obtaining improved coke by selectivity preparing and blending coals - particularly coals that do not have the proper petrographic composition - for producing the most satisfactory coke.

The petrographic components of another high-volatile bituminous coal were concentrated by gravity separation in liquids of adjusted densities. Chemical analyses of the concentrates showed considerable differences in chemical composition. X-ray scattering patterns indicated that, although these components have the same basic aromatic layer structure, they appear to have different distributions of amorphous carbon and groups of aromatic layers of different diameters. Higher contents of amorphous carbon and smaller size groups are characteristic of exinite and vitrinite, whereas larger layer groups predominate in micrinite and fusinite.

[15/] Parks, B. C., O'Donnell, H. J., and Darakos, W. E., Relation Between Petrographic Composition and Free-Swelling Properties of Chilton Coal: Bureau of Mines Rept. of Investigations 5294, 1957, 22 pp.

A summary was presented of the Bureau's thin section petrographic work from its beginning to 1956.[16]

Mild Chemical Treatment

Studies of coal structure have been hampered by lack of a method for reductive degradation of coal at low temperatures. Metals in liquid ammonia reduce many organic sutstances, including lignin; however, the insolubility of coal in ammonia renders this reagent useless. Lithium in ethylenediamine, a good coal solvent, was found to be effective in reducing phenols and polycyclic aromatic hydrocarbons.[17] Four coals of varying ranks and a pyridine-soluble extract of one of them were reduced by lithium in ethylenediamine at 90°-100° C. Comparison of the infrared and ultraviolet spectra of the reduced coals with those of the untreated coals showed direct addition of hydrogen to carbon, a decreased number of oxygen linkages, and decreased aromatic structure. The pyridine extract showed further reduction than the coals. This reagent reduced even an anthracite, its hydrogen content increasing 1.3 percent by weight.

Researchers studied another low-temperature hydrogenation reaction: Two coals were treated with an equimolar mixture of $CO+H_2$ in the presence of dicobalt octacarbonyl at 200° C. Infrared analysis of the products showed that carbonyl and hydroxyl groups had been added and hydrogenation had occurred. This is the first example of the addition of molecular hydrogen to coal at temperatures as low as 200° C. These experiments also indicated the presence of nonaromatic double bonds in coal. Optimum conditions for reducing and solubilizing coal are being investigated to provide samples for an intensive characterization study.

Previous work has shown the merits of hexamethyldisilazane, $(CH_3)_3SiNHSi(CH_3)_3$, for quantitative determination of hydroxyl groups. More than 20 coals of varying rank have been treated with this reagent and the resulting silyl ethers analyzed for silicon. From the silicon analyses the percentage of oxygen present in the coals as hydroxyl groups has been calculated. Plots of hydroxyl content as a function of rank or of carbon content indicate that for coals of carbon content up to about 80 percent the hydroxyl groups are approximately 5 percent by weight of the coal; above 80-percent carbon content, the percentage of hydroxyl groups steadily decreases to zero at about 94 percent carbon. The ratio of oxygen present in hydroxyl groups to the total oxygen content shows a maximum value of about 50 percent for a coal of about 81 percent carbon; this ratio decreases continuously to values of about 10 percent for lignite and anthracite.

Humic Acids

Humic acids, essentially a class of hydroxy carboxylic organic acids available from oxidized coal or humic soils, are readily obtained from naturally or artificially oxidized lignite, and their investigation may serve two purposes: Ultimate utilization of the acids as such or as intermediates and their use in solving problems of coal structure and coalification. Samples of oxidized lignite from large deposits in southwestern North Dakota, locally known as leonardite, were extracted with mild alkali, dissolving essentially all but the inorganic mineral content. This alkali-soluble material, recovered by acidifying the caustic solution and collecting the precipitated acids, was fractionated by extraction with various organic solvents. This work is continuing, and the object is to further simplify fractions so that basic units of the original acid complex may ultimately be identified.

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[16] Parks, B. C., and O'Donnell, H. J., Petrography of American Coals: Bureau of Mines Bull. 550, 1956, 193 pp.

[17] Reggel, L., Friedel, R. A., and Wender, I., Lithium in Ethylenediamine; a New Reducing System for Organic Compounds: Jour. Org. Chem., vol. 22, No. 8, August 1957, pp. 891-894.

Pulverized samples of a "normal" lignite were oxidized in air at 100°-150° C., and the product was extracted as in the case of leonardite. This material appears to be quite similar to the soluble portions obtained from naturally occurring leonardite. On the other hand, H:C and O:C ratios differ somewhat between leonardite and oxidized lignite, indicating the possibility of chemical structure differences between the two materials.

High moisture-holding capacity is characteristic of both humic acids and lignite, and a paper has been published on determination of moisture in lignite.18/

Microbial Degradation

Further studies of bacteriostatic and fungistatic materials in coals have shown that extraction of the active portions of methanol-soluble material with water may increase the activity roughly three- to five-fold. A five-fold increase in activity brings the potency of the material to within 1/300 to 1/500 that of the antibiotic penicillin. A large sample of methanol-soluble material has been obtained by treatment of a 20-pound coal sample; an attempt will be made to concentrate the active principle from this material.

The best growth of organisms on coal to date has been found in two mold cultures isolated from an Illinois coal from the Eddy Mine. One of the cultures is an *Aspergillus*; the other is as yet unidentified. The cultures have been carried through a number of transfers in a medium containing 4 percent of the coal as the sole carbon source, and growth has been maintained with no nutrient additions to the medium.

Additional studies on the oxidation of polycyclic aromatic compounds19 20/ by bacteria confirm the hypothesis that different enzymes are formed in response

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18/ Fowkes, W. W., Hoeppner, J. J., and McMurtrie, R., Some Notes on the Determination of Moisture in Low-Rank Fuel (Lignite): Fuel, vol. 36, No. 4, October 1957, pp. 469-474.

19/ Rogoff, M. H., and Wender, I., The Microbiology of Coal. I. Bacterial Oxidation of Phenanthrene: Jour. Bacteriology, vol. 73, No. 2, February 1957, pp. 264-268.

20/ Rogoff, M. H., and Wender, I., 3-Hydroxy-2-Naphthoic Acid as an Intermediate in Bacterial Dissimilation of Anthracene: Jour. Bacteriology, vol. 74, 1957, pp. 108-109.

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to linear polycyclic aromatic hydrocarbons (anthracene) as opposed to angular polycyclics (phenanthrene).

Irradiation of Coal

Five coals subjected to pile irradiation by the Federal Geological Survey have been investigated by infrared and by electron resonance. The greatest change in these irradiated coals was a pronounced increase in hardness. The infrared spectrum for each irradiated coal was more diffuse than that of the nonirradiated coal, but the characters of the spectra were identical. The results indicate that the apparent rank of the coal had not been changed, as had been postulated from changes in carbon-hydrogen ratios. Free-radical contents of these five coals were investigated before and after pile irradiation. The two highest rank coals (about 90 percent carbon) showed a five-fold increase in free-radical content, but the lower rank coals showed no increase.

A small amount of soluble material extracted from a coal subjected to gamma radiation in a carbon tetrachloride slurry was obtained from another laboratory. The infrared spectrum of this product differed considerably from the spectrum of the original coal.

A book on the chemistry and physics of coal was reviewed.[21/]

EXPLORATION AND MINING

Exploration of Alaska Coals

The Bureau of Mines is cooperating with other Federal agencies in an effort to develop a coal-mining industry in Alaska to supply the increasing solid-fuel requirements for both military and civilian installations. The work includes trenching and diamond core drilling to determine the position, size of reserves, and quality of coalbeds where these data are not known.

Both churn- and core-drilling were continued throughout the 1957 field season in the western part of the Wishbone Hill district, Matanuska coalfield (fig. 1). This coalfield has been studied for many years and contains both bituminous and subbituminous coal.[22/] The objective of the drilling was to obtain data on the location and character of the coal measured. Several beds intersected and correlation of their location with known series of the beds is in progress.

The Bureau of Mines conducted a reconnaissance examination of the less accessible parts of the Beluga coalfield in August 1957 (fig. 2). This coalfield, 50 (airline) miles west of Anchorage, is of increasing interest as a possible source of large tonnages of strip coal for on-site production of power.

[21/] Howard, Henry, C., Book Review of Coal Science: Scientific Monthly, vol. 85, No. 5, November 1957, pp. 278.

[22/] May, R. R., and Warfield, R. S., Investigation of Subbituminous-Coal Beds Near Houston, Westward Extremity of Matanuska Coalfield, Alaska: Bureau of Mines Rept. of Investigations 5350, 1957, 20 pp.

Technologists examined 13 separate coal outcrops from which 8 samples were taken. The coal is of subbituminous rank. Further investigation will be necessary to determine the extent of possible strippable coal.

Anthracite Mining

Longwall Mining

Since 1945 mining engineers have initiated and conducted experiments for planing coal and for a roof-support system (yielding steel props) applicable to longwall mining. The results of experiments with the Bureau-designed pneumatic coal planer have been published.[23] In the Northern anthracite field, where continuous miners, universal shearers, mobile loaders, and shuttle cars cannot be used in seams with a pitch greater than 10°, full mechanization of the longwall caving system might be applied to recover the millions of tons of coal in seams with pitches from 10° to 20°. A cooperative agreement has been made to test this method in an operating mine. The block of coal to be mined in this experiment is approximately 1,200 feet in length, providing for 200 days of operation with an average advance of 6 feet per 24-hour day.

Scraper-Shaker Loader

Several years ago the Bureau designed a scraper-shaker loader employing the relatively low-cost, low-maintenance scraper in combination with a shaker conveyor for developing gangways in thin (4 to 8 feet thick), steeply pitching (60° or more) beds. Investigations disclosed a location in the Western Middle field of the Anthracite region where a new rock slope was to be driven from the surface to develop a new section of a mine in which the machine could be tested. In this test the machine was modified by removing the shaker drive and rear suspension bridge, and the slope was driven 200 feet, the limit originally established for penetrating the bed to be mined. The scraper-shaker loader removed blasted material in one-fourth the usual time, with one-fourth the number of men - a sixteenfold improvement over conventional hand-loading methods.

Mine-Drainage Control

A joint Federal-State program for controlling water in anthracite mines was initiated in 1955 under enabling legislation of the Congress of the United States and of the General Assembly of the Commonwealth of Pennsylvania. A total fund of $17 million was provided for the installation of surface drainage works, pumping works, and related facilities for controlling and draining water, which, if not so controlled or drained, would cause flooding of anthracite formations. Two projects were approved in 1956, and an additional 14 were approved in 1957. The first project to be completed under the program was installation of concrete flumes and pipes in the stream bed of Boston Creek in Luzerne County (fig. 3). The need for this improvement has been shown by the fact that water 6 to 9 inches deep now flows through the flume for several days

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[23] Buch, J. W., Griffith, W. R., and Schimmel, J. T., Design and Testing of Bureau of Mines Pneumatic Coal Planer: Rept. of Investigations 5380, 1957, 25 pp.

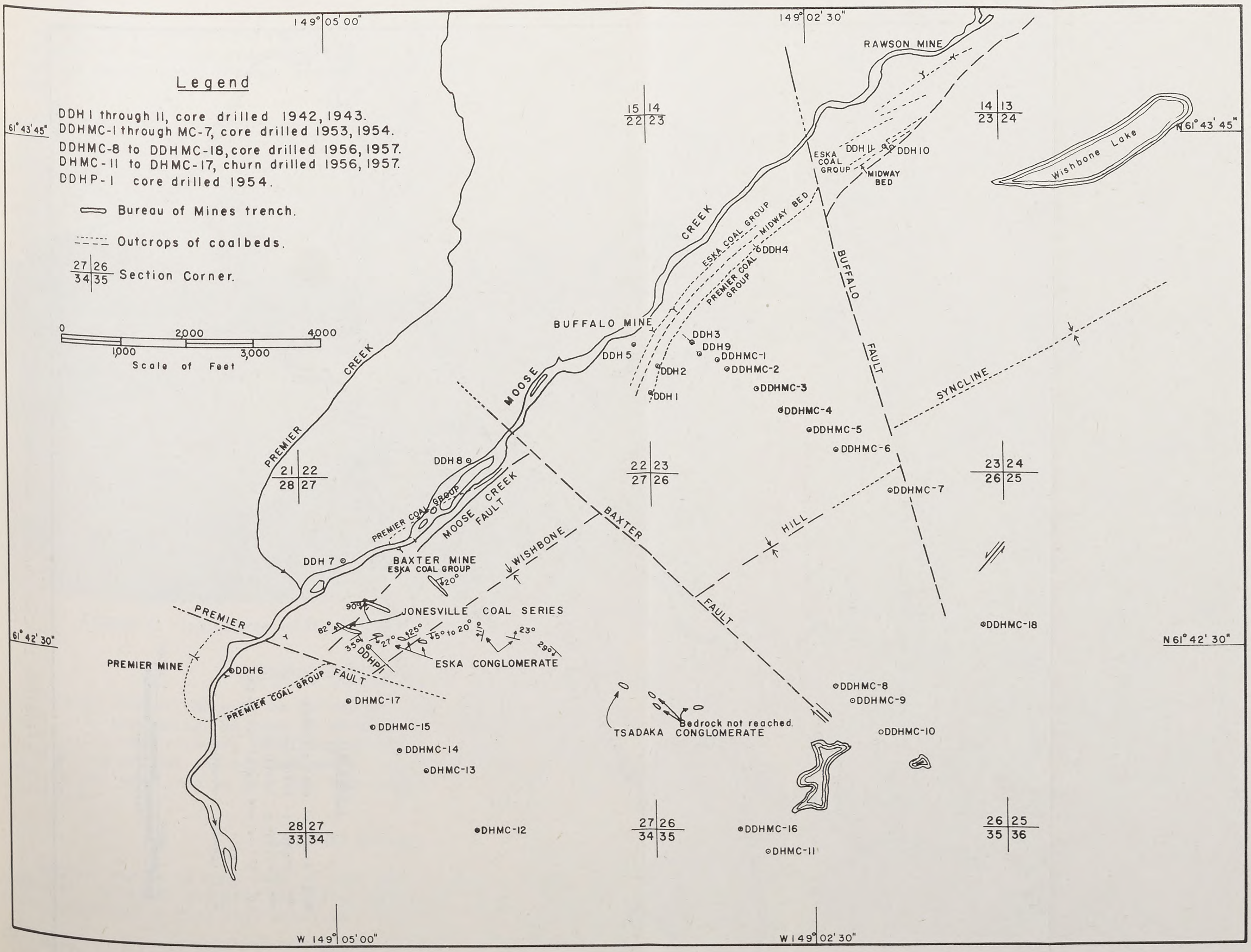

FIGURE 1. - Western Part of Wishbone Hill District, Matanuska Coalfield, Alaska.
(Modified from Map of U.S. Geological Survey.)

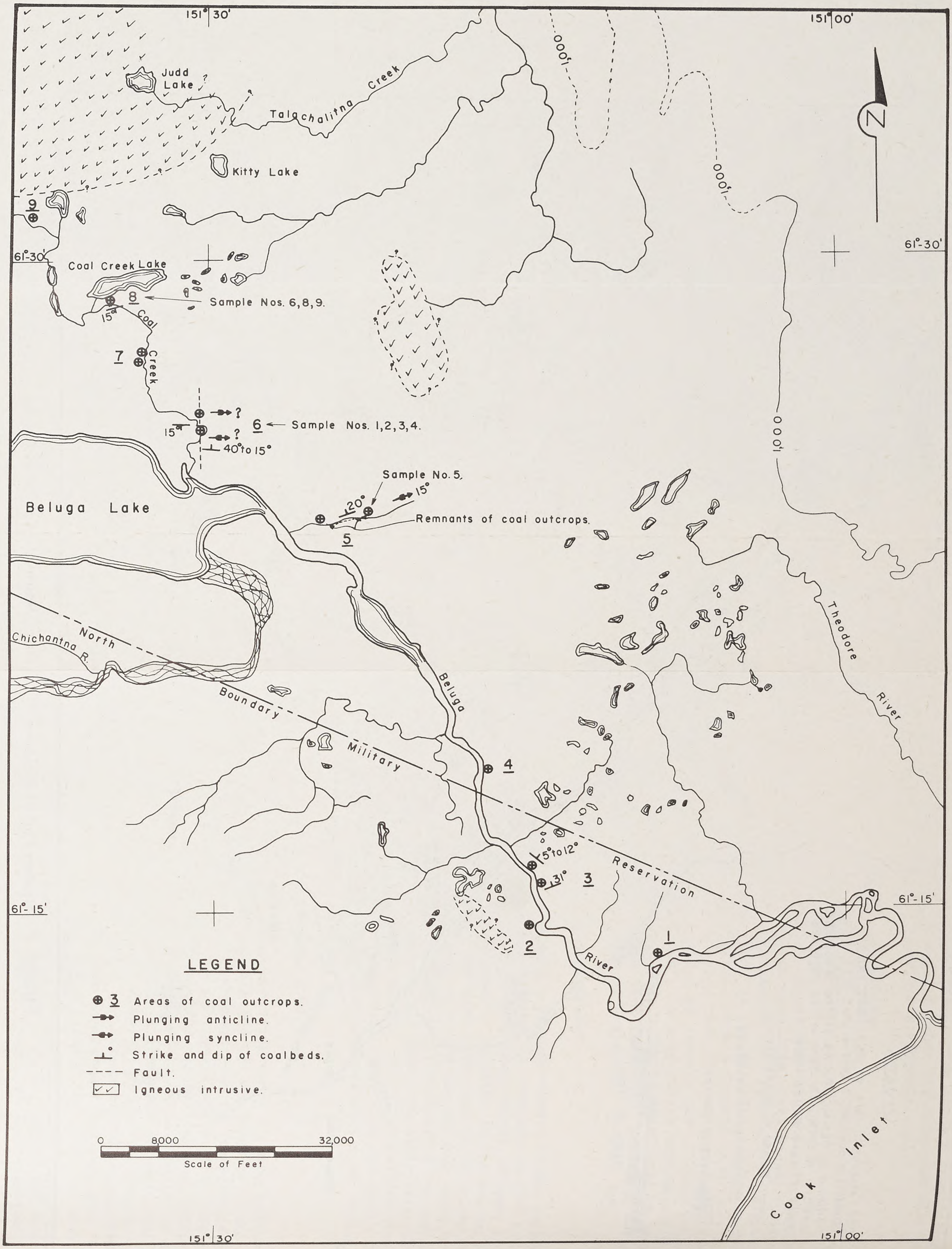

FIGURE 2. - Coal Outcrops Near Beluga Lake, Alaska.
(Modified from Map of U.S. Geological Survey.)

after a heavy rain. Before this project was constructed there was virtually no flow of water in the creek bed after heavy rains because all the surface water reaching the creek seeped through broken rock strata into the underground workings.

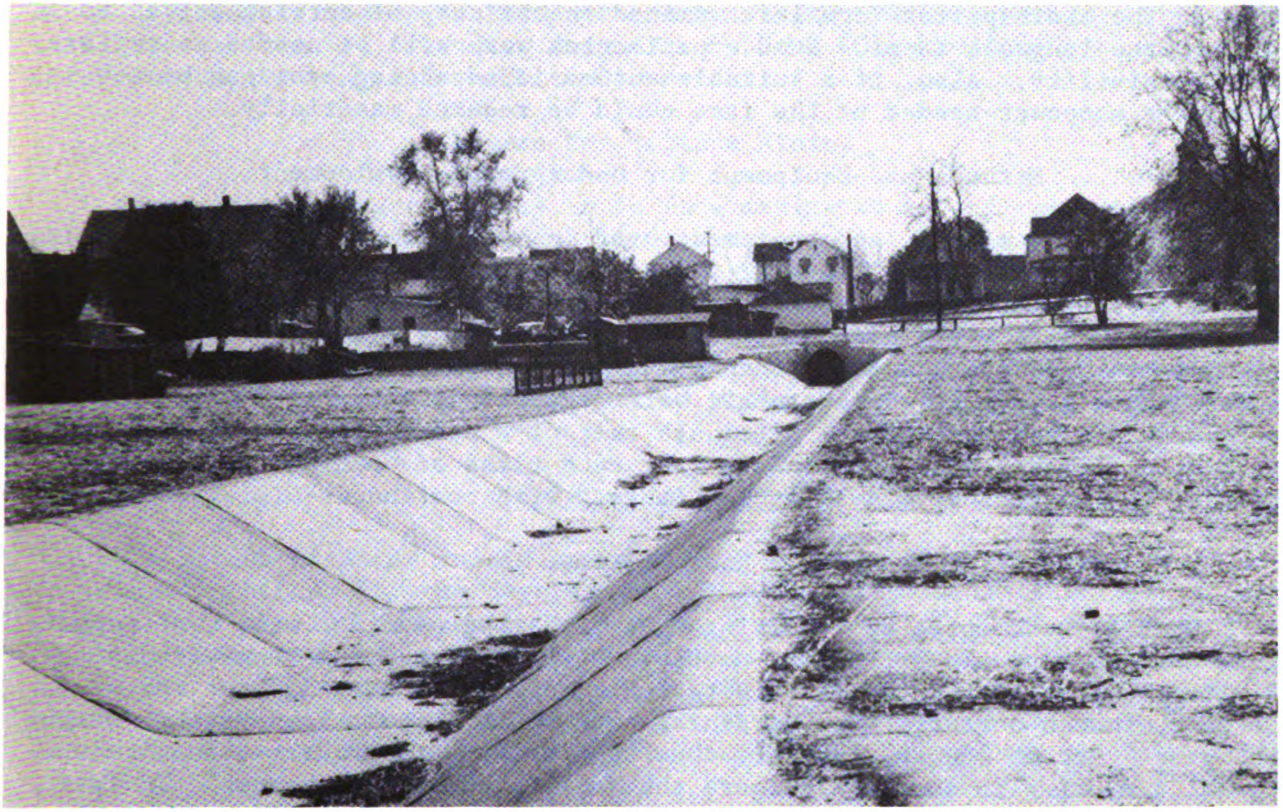

FIGURE 3. - Improvement of Stream Bed, Boston Creek, Larksville, Pa.

Bituminous Coal Mining

The objective of the work on bituminous coal is to develop improved methods for: (1) Increasing the rate of production to reduce the cost of coal and (2) increasing coal recovery in mining to conserve natural resources. Data on various mining methods and performance of equipment are collected, compiled, evaluated, and published so that the results for given operating conditions can be compared.

Longwall Mining

Longwall mining of thin beds of coal with a German coal planer has proved successful. A report has been published summarizing, to June 30, 1957, the planer operations at five coal mines.[24] When the publication was prepared,

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[24] Haley, W. A., and Dowd, J. J., Modified Longwall Mining With German Coal Planers. Summary of Operations at Five Coal Mines: Bureau of Mines Rept. of Investigations 5355, 1957, 31 pp.

Original from THE OHIO STATE UNIVERSITY

one of the planers had been taken out of service because the coal being mined was too hard. Another planer was just being placed in operation but had not produced any coal. The total production to the end of 1957 was 2,381,546 tons.

Recovery of coal by the longwall method can be increased by recovering some of the chain pillars now left unmined in the sets of entries driven to develop the longwall faces. Some experimental work will be needed to explore this possibility. Also, if a suitable automatic advancing prop can be developed, the manpower needed at the face could be reduced materially.

Methods and Equipment for Underground Development

A report on methods and equipment used in underground development by bituminous coal mines was published.[25] A study of 14 mine operations shows that modern development methods permit close supervision owing to close proximity of the working faces. Efficient use of machinery is possible because long moves of equipment are unnecessary. Each member of a development unit can operate each machine used, and therefore all work together as a team. The width of development openings approaches or equals that of a production place, and there is little difference in the tonnage from each class of opening. As was expected, the output per man-shift was largest with continuous-mining machines.

Roof Bolting

Studies to compare roof bolting with timbering used for mine roof support were continued at mines in Ohio, northern West Virginia, and Pennsylvania. Fieldwork is still in progress. Detailed studies have been made at two mines in Ohio, two in northern West Virginia, and one in western Pennsylvania during the year. Recent studies indicate that roof bolting is preferred over conventional timbering in some mines where the roof is considered tender. In fact, some mines use roof bolts in conjunction with timber to support tender roof.

Systems and Equipment for Face Haulage

The introduction of continuous-mining machines created a new haulage problem. The coal could not be moved away from the machines fast enough. This resulted in lost time for the mining machine until the mined coal could be moved. Systems of face haulage and equipment used in this phase of mining are being studied to determine the best systems of face haulage. Studies have been made in 12 mines in which various combinations of methods and equipment can be used. At each mine where more than one system is in use they are compared to determine the relative efficiency.

Recovery of Coal

The percentage of coal recovered in mining varies with the coal beds and methods of mining. In mines where pillars are not recovered and no complete record of production is available the calculated recovery must be based on the measured area of the remaining pillars, as shown on the map of a mined area. Where pillars are recovered, the percentage of recovery must be calculated from production records. These records are not always obtainable due to incomplete records or transfer of ownership. Recoveries have been calculated for five mines operated in the Upper Freeport and two in the Pittsburgh bed. Where pillars were not removed, recovery ranged from 50 to 60 percent; where pillars were removed, it ranged from 80 to 88 percent.

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[25] Shields, J. J., Dowd, J. J., and Haley, W. A., Mechanical Mining in Some Bituminous-Coal Mines. Progress Report 8. Methods and Equipment Used in Underground Development: Bureau of Mines Inf. Circ. 7813, 1957, 66 pp.

Augers for Surface Mining

Augers are used principally in surface mining to recover coal that cannot be mined by conventional methods. With augers, coal can be recovered near the outcrop under top that is too weak for underground mining or under too much cover to be strip mined. Auger mining requires only a few men, and the output per man-shift is larger than that from either underground or strip mining. In a recent publication the operation at eight mines revealed that, although coal can be produced cheaply by this system of mining, the recovery in mining ranges from 20 to almost 50 percent.[26]

Foreign Activities

The Bureau of Mines continues to support a program of technical assistance to friendly nations for the development of their solid fuels resources in cooperation with the International Cooperation Administration, U. S. Department of State. Inadequate health and safety measures and a shortage of trained personnel for mining operations are problems of greatest concern to Bureau engineers assigned to technical assistance posts in Afghanistan, Colombia, and Indonesia, where coal production must be increased to encourage the industrial growth of these countries.

Bureau facilities and personnel in the United States support the work of the field staff by laboratory research and by training foreign nationals in all subjects regarding exploration, mining, and utilization of coal. Eleven foreign engineers were accepted during the year for training at Bureau facilities. In addition a Bureau team of health and safety engineers conducted a 4-month training course in mine rescue and first aid for 587 employees at Mexican mines to improve standards of safety in that country.

Afghanistan

The lack of mining timber in Afghanistan has retarded development and production at the country's two producing coal mines, Karkar and Ishpushta, where bad roof conditions add hazards to generally unfavorable mining conditions. Tree gardens must be planted as part of all mining operations, and past experience shows that a 10- to 12-year growth is required to produce softwood suitable for underground roof supports. The mining companies have acknowledged the success of these experiments, and larger areas of irrigated land are being acquired for cultivating approximately 100,000 trees that will be needed for future mine operations.

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[26] Haley, W. A., and Dowd, J. J., The Use of Augers in Surface Mining of Bituminous Coal: Bureau of Mines Rept. of Investigations 5325, 1957, 22 pp.

The Afghan Government has purchased 100 American trucks for transporting increased tonnages of coal from the Karkar and Ishpushta mines to industrial and domestic markets. A Swiss briquetting machine is now being assembled in Kabul. Degradation in handling and shipment of the friable coal has limited its use as domestic fuel; to increase domestic consumption, briquets will be made from low-grade coal mixed with ground cottonstalks as a binder.

Completion of a powerline between the mine and the hydroelectric plant at Pul-i-Khumri improved ventilation at the Karkar mines by permitting electrification of mine fans. Modest improvements have been made in living accommodations for the workers at the mine, and health and safety centers at each mine have been provided with medical equipment and medicines. Mine-accident and minor surgery cases can now be handled promptly, and the patients are given post-surgery care for a few days at the health centers.

Development of the Darra-Suf coalfield is still in the planning stage and will not be started until a decision is reached regarding the location of a road needed for transporting coal.

Colombia

A reported reserve of over 200 million tons of coal in the El Valle and Cauca areas of Colombia can be the basis of a substantial and profitable coal industry if the many small and poorly equipped mines in the area are organized under some plan whereby a dependable source of export-quality coal can be established and an anticipated export trade can be realized.

A new coal washing and preparation plant was erected near Cali, in the Cauca Valley, by the Instituto de Fomento Industrial, a Colombian Government corporation responsible for industrial development. The plant operated throughout the year; although its mechanical performance has met all expectations in respect to the quality of coal produced, its potential benefits to the coal industry are not yet fully realized. The plant was built primarily to prepare coal of controlled quality for export markets. Plant operators purchase mine-run coal, and payments depend upon the percentage of mineral matter in the coal. On an average, the price paid at the washing plant is approximately $0.50 per ton less than the local market price, hence the tonnage delivered is far below the rated capacity of the plant.

One shipment of approximately 10,000 tons of washed coal was sent to France late in the year, but export shipments probably could be increased to more than 100,000 tons a year. Coal reserves, coal-preparation facilities at Cali, and transportation facilities for delivering coal to the docks at Buenaventura are all adequate for this tonnage. The principal need is financial and technical assistance for the small mine operators. Preliminary steps have been taken to provide financial assistance through loans to independent operators for purchasing equipment, and the technical assistance project has been extended to continue the advisory service, particularly in mine safety and mechanization, now being given by a Bureau of Mines engineer.

Indonesia

The Bureau assigned a coal-mining engineer early this year to Indonesia to assist the Indonesian Government in a development and expansion program for the coal industry, which, in recent years, has experienced a serious deterioration in efficiency.

Major coal production in the past has been from the Ombilin mine in central Sumatra and from the Bukit Asam coal mine in southern Sumatra. Present endeavors are aimed at reestablishing the Ombilin mine in its former position as a leading coal producer. Total reserves in range of present mine openings exceed 40 million tons of coal occurring in two main seams. Present production from underground mining operations is about 225 tons daily or approximately 75,000 tons a year. A newly established production schedule calls for an annual output of 300,000 tons, which should be attainable without an appreciable increase in the labor force.

The introduction of strip mining at the Ombilin mine has been proposed for one area and should yield approximately 500 tons of coal per day. Rehabilitation of underground workings and improvements in mining operations are expected to increase mine production to approximately 1,000 tons a day.

Seventy percent of the current coal production in Indonesia is from the Bukit Asam coal mine. The present production at this mine, amounting to approximately 20,000 tons a month, is by strip mining. A study is being made to assess possibilities of increasing production to supply part of the 18,000 tons of coal a month that will be required for a proposed powerplant at Surabaja.

The Bureau's engineer has prepared and submitted reports outlining procedures whereby production can be increased and improvements made in health and safety practices at the mines.

India

A Denver engineering firm, under the direction of Bureau engineers, has completed the design and fabrication of all major units for a complete pilot plant to crush, dry, briquet, and carbonize lignite. This pilot plant has been shipped to the Neyveli Lignite Corporation and will be erected at Neyveli, Madras State. The plant should be operating in May or June 1958; two Bureau engineers will then go to India to supervise initial operation and to work with Indian technologists in interpreting operating data and to assist in working out a program of further research directed toward utilization of lignite. Further details will be found under Utilization of Lignite From India, page 59.

Health and Safety

Roof Control

The expense of roof support ranks as one of the major items in the production cost of bituminous coal. To a considerable extent, roof bolting has offset this increase in cost by providing more unobstructed operating space than is provided with conventional timbering. Long haulways, airways, and unattended belt haulage systems have focused attention on the necessity of providing a stable roof to give greater security, continuity of operation, and a lower mine maintenance cost.

Roof bonding, or solidification of ground has been studied during the past year. Initially, experiments were conducted with oils to determine the extent of penetration at various pressures in different types of roof rock. Epoxy resins of various viscosities were used in later experiments as bonding agents to determine penetration, bonding qualities, and setting. Indications are that resins can be injected into strata and that such resins will effectively bond pieces of rock. Important goals in future experimental work are to design mining and pumping equipment, to discover a low cost bonding agent, and to develop a systematic, economic procedure.

Roof bolting continued its growth during the year. More than 50 percent of the underground bituminous coal is mined under bolted roof. Roof-bolt failures caused less than 2 percent of the roof-fall fatalities. Bureau records show that approximately 80 percent of all roof-fall fatalities in bolted areas occurred beyond the point where the bolts were installed.

Determination of roof stability, under bolted conditions, is one of the remaining complex problems, due to the variations of rocks that make up mine roof and the diversity of mining systems. This problem is associated, to some degree, with the extent and time of roof sag and with the change in bolt load. Significant data have been collected in mines operating in the Pittsburgh coal bed where test zones of varied bolting patterns have been established. In other zones slotted-type and expansion-type shells are used under similar conditions to determine their comparative effectiveness in stabilizing roofs.

New developments being studied include: (1) An expanding rubber sleeve to provide more anchorage in softer shales; (2) a self-indicating tension device, which is a headed bolt with a crimped flash that flattens at a predetermined pressure; (3) a penetrometer to measure the hardness of rock in drilled holes; and (4) a mining shield to protect miners and other underground workers from the hazards of falling rock and other materials.

Roof-Fall Fatalities

During 1956, 214 fatalities from falls of roof, face, or rib were investigated, and the circumstances and causes of the accidents were reported.27/ About 63 percent of all underground fatalities in bituminous coal and lignite mines resulted from such falling material, compared with 65 percent for 1955; however, the fatality rate of deaths per million man-hours from this cause was 0.58 in 1956, as compared with 0.54 in 1955 and 0.52 in 1954. Human failure was considered responsible for 96 percent of these fatalities, and only 4 percent were charged to circumstances or risks that apparently could not be

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27/ Joseph, R. O., and Thomas, E. M., Falls of Roof. The No. 1 Killer at Bituminous-Coal Mines, 1956: Bureau of Mines HSS 454, Mineral Industry Surveys, 1957, 13 pp.

foreseen. Seventy-five percent of the fatalities resulted from accidents within 25 feet of the working face, and 52 percent of the fatalities in this area occurred between the last permanent roof support and the working face. The average length of the comparatively small area from the face to the last permanent support - the most dangerous part of a mine - was 9 feet.

The average dimensions of the pieces of roof rock that fell and resulted in fatalities were: Length, 13 feet; width, 12 feet; and maximum thickness, 2 feet. The relatively large area of these falls shows that adequate minimum standards for roof control should be adopted and followed in all mines. Coal was loaded mechanically in places where 69 percent of the face fatalities occurred; the remaining 31 percent of the fatalities occurred in places where the coal was hand-loaded.

Twenty-four fatalities occurred in places where roof bolts were used, which represent 11 percent of the total roof-fall fatalities. In only four of the falls did roof-bolt failure cause the accident. In most of these accidents the fall occurred inby the bolted area; and temporary roof supports, such as safety posts, had not been installed. Roof bolts generally offer an improved method of roof control but do not obviate the necessity of providing temporary support inby the bolted area. Indications are that the fatality rate, based on tonnage, from falls of roof in bolted areas is less than that in areas where conventional timbers are used.

Reports on 48 underground fatalities in the Pennsylvania anthracite mines in 1956 revealed that roof falls were responsible for about two-thirds of the underground deaths.[28] Haulage accidents caused one-eighth of the fatalities. The remainder were due to strip-mine accidents, electricity, and other causes. The responsibilities of management and employees in preventing fatalities from roof falls were discussed, and suggestions to assist in accomplishing this goal were presented.

Haulage Fatalities

During 1956, 82 haulage fatalities occurred at underground bituminous coal mines.[29] Of these, 71 occurred underground and 11 on the surface. Haulage accidents at strip mines caused nine fatalities but are not included in the report. Exactly one-half of the reported haulage fatalities were in West Virginia. Alaska and 17 coal-producing States were free of such fatalities. Of the 82 fatalities, 36 occurred in violation of some section of article VII of the Federal Mine Safety Code.

Some form of physical or operating hazard was considered to be the primary cause of 54 percent of the fatalities. Unsafe practices, either involuntary acts or deliberate chance taking, were considered to be the primary cause of 40 percent of the fatalities. The Bureau analyzed the responsibilities of management and employees with relation to safe haulage operations and presented recommendations to reduce the frequency of haulage accidents.

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[28] Mather, J. V., Fatalities at Pennsylvania Anthracite Mines, 1956: Bureau of Mines HSS 452, Mineral Industry Surveys, 1957, 24 pp.

[29] Smith, G. M., Haulage Fatalities in Bituminous-Coal Mines, 1956: Bureau of Mines HSS 453, Mineral Industry Surveys, 1957, 13 pp.

Coal-Mine Injury Data

A summary and analysis of reports from coal-producing companies and State mine officials on injuries and related employment data in the coal-mining industry for 1952 through 1955 showed a progressive and gradual improvement over the preceding years. Incomplete reports (those received in the Washington office by June 15, 1957) for the year 1956 showed a slight upward trend in frequency of injuries as compared with 1953, 1954, and 1955, although slightly lower than in 1952. The lowest number of deaths to occur in coal mines in the United States was in 1954 when, for the first time since statistics have been collected by the Bureau of Mines, the death toll dropped below 400 in a year. In 1952 there were 2 major disasters (single accidents in which 5 or more men are killed) which claimed the lives of 11 men; in 1953 there was 1 in which 5 men were killed; and in 1954, 1 in which 16 were killed. From November 13, 1954, no major disaster occurred in the industry through 1956 - a period of more than 2 years.

In 1956, 444 men were killed in accidents at bituminous coal, lignite, and anthracite mines at frequency rates of 0.84 per million tons of coal produced and 1.08 per million man-hours of time worked.[30] Although 27 more men were killed at coal mines in 1956 than in 1955, the rate of 0.84 per million tons remained the same as in 1955. The rate per million man-hours worked increased from 1.04 to 1.08. The increase in the number of fatalities in 1956 occurred in underground mines. Sixteen more men were killed by falls of roof, face, or rib in 1956, and underground haulage accidents claimed the lives of 11 more men than in the preceding year. Surface operations at deep mines showed a decrease of 14 fatalities and strip operations had 1 more fatality. The coal mines in the United States were free of a major disaster (one in which 5 or more men are killed) during 1956. The last major disaster occurred in a West Virginia mine on November 13, 1954, when a gas explosion resulted in the death of 16 men.

Safety in Shaft Sinking

Unfortunate occurrences resulting in injuries, loss of life, and destruction of property by explosion, fire, and other mishaps during shaft sinking in the past few years have focused the attention of the Bureau of Mines on the need for safety standards for such operations.[31] One hundred and eighty safety standards have been assembled under the following main categories: Fire prevention, fire protection; housekeeping and sanitation; electricity; explosives storage and transportation; timbering - safeguarding personnel and equipment; hoisting, hauling, and handling excavated material; ventilation; drilling; blasting, welding and burning; and lighting. Bureau personnel has compiled the recommended standards included in this publication in all areas where shaft sinking is practiced. Existing applicable State rules were considered and included. Representatives of the mineral industries, coal and noncoal, were given an opportunity to participate in preparing these suggested standards.

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[30] Reid, Elizabeth J., and Wrenn, Virginia E., Coal-Mine Fatalities in 1956: Mineral Industry Surveys CMF 306, 1957, 9 pp.

[31] Recommended Safety Standards for Shaft Sinking: Bureau of Mines Inf. Circ. 7810, 1957, 21 pp.

Mine Rescue Equipment

In recent years the Bureau of Mines has tested and approved four new types of self-contained breathing apparatus and a facepiece designed for use with the McCaa 2-hour self-contained oxygen breathing apparatus.32/ The breathing apparati are 1/2- and 3/4-hour self-contained types of much simpler design than the conventional 2-hour unit ordinarily used by mine rescue crews. The new equipment is approved only as auxiliary equipment; the 2-hour, conventional-type apparatus should be used for training.

Communication

Recent developments in portable communication equipment for use in emergency recovery work following mine fires and explosions have made possible a convenient and dependable means of directing rescue teams working under hazardous conditions.33/ Mine rescue and recovery operations following fires and explosions are frequently conducted under adverse conditions that require the use of oxygen breathing apparatus by members of a five- or six-man team. The use of conventional apparatus presented difficulties in developing telephone communication because of the wearer's inability to speak while wearing the mouthpiece. The report cited discusses development of communication equipment and devices, from the sash-cord lifeline to the recent transistor-operated telephone.

Trailing Cables

As a result of research suggested by the Bureau of Mines, a cable manufacturer has developed an incombustible, thermoplastic trailing cable that has satisfactorily passed the flame and damage tests for fire-resistant cables under Schedule 2F, Regulations Governing Permissibility Tests for Electric Face Equipment. Favorable reports from field tests of these cables indicate that the durability of these cables in actual service exceeds that of some fire-resistant cables previously available to the industry. Acceptance of these cables by the mining industry should greatly reduce the frequency of cable failures - the most serious mine-fire hazard confronting it.

The reduction of effective insulation and the increased rigidity of trailing cables necessitated by incorporating an additional conductor for frame grounding have caused many trailing-cable failures, some with disastrous results. Through research by the Bureau of Mines and the cooperation of

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32/ Walker, W. Dan Jr., Chastain, G. W., and Dornenburg, D. D., Auxiliary and Supplemental Mine Rescue Equipment: Bureau of Mines Inf. Circ. 7808, 1957, 50 pp.

33/ Brown, C. L., Development of a Transistor-Type Telephone System for Mine Rescue Operations: Bureau of Mines Rept. of Investigations 5318, 1957, 11 pp.

manufacturers improved equivalent protective devices to replace frame-grounding conductors have been developed and certified by the Bureau of Mines as acceptable. Personnel of the Bureau is making field surveys to determine the effectiveness of these devices in actual service.

Mine Lighting

During 1957 the Bureau announced permissive approval of four generalized mine-lighting systems under Schedule 29, Mine Lighting Equipment for Underground Workings. Encouraging the use of these approved mine-lighting systems and continuing research to improve them should aid in overcoming one of the greatest handicaps to safety and production of underground works, that of poor visibility. Reduction in injuries and increased production are expected to result from the use of approved mine-lighting systems.

Documentary evidence at disasters and at sites of other unusual underground accidents can now be obtained with an explosion-proof, high-power floodlamp and development of a complete system with which these lamps may be safely used. The unit was tried successfully for the first time in September 1957, when scenes were photographed after a coal-mine disaster at Marianna, Pa.

Mine Ventilation

Studies have been continued of major ventilating problems confronting the coal-mining industry. Face ventilation with continuous-type mining machines has been given particular attention. Research is being conducted to develop methods of automatically cutting off power to machines in face areas when the percentage of methane reaches a predetermined minimum and also to develop an audible warning device to alert workers that the concentration of methane is approaching the danger point.

The absolute pressure method of making ventilation surveys in coal mines defines areas of above-average resistance to airflow, and details of the operational procedure were published.34/ An electric analogue, developed for solution of mine ventilation network problems, has proved valuable in many instances.

As part of the program of coal-mine inspection under the provisions of the Federal Coal Mine Safety Act, approximately 19,000 samples of mine air were analyzed to determine the adequacy of ventilation in coal mines, to detect and aid in eliminating hazards from flammable and toxic gases in mines, and to provide information necessary in controlling and extinguishing fires in coal mines. In the analysis of samples of coal-mine atmosphere conventional gas-volumetric analysis methods were supplemented by the use of infrared absorption spectrometry to determine, both qualitatively and quantitatively, the methane content of certain samples of particular importance.

34/ McElroy, G. E., and Kingery, D. S., Making Ventilation-Pressure Surveys With Altimeters: Bureau of Mines Inf. Circ. 7809, 1957, 20 pp.

Dust Control in Mines

Three drill dust collectors for coal-mine use were approved during 1957, bringing the total of approved collectors to 46. Numerous minor modifications in previously approved equipment were covered by extensions of approval. Dust control in coal mines was discussed at a symposium on coal workers' pneumoconiosis,[35] and information was presented on concentrations of airborne dust resulting from representative mining operations in several bituminous coal mines with different methods of dust control in effect.[36]

Safety Education

In the 1957 National First-Aid and Mine Rescue Contest, Louisville, Ky., 57 teams from 7 States participated - 8 teams from 3 States in the mine rescue contest and 49 teams from 7 States in first aid. The contest was held under the auspices of the Federal Bureau of Mines and the Joseph A. Holmes Safety Association and was sponsored by the National Coal Association, United Mine Workers of America, and various State mining departments.

STORAGE AND PREPARATION

Storage of Lignite

Stable, long-term storage of lignite has been demonstrated at Garrison Dam, Riverdale, N. Dak., where over 2 million tons of lignite excavated during construction of the dam, has been stored. Bureau of Mines personnel in cooperation with personnel of the Army Corps of Engineers developed the method of stockpiling. Piles constructed in 1-foot lifts and compacted by sheeps-foot rollers are preserved in their initial condition. Erosion of the protective slack layer by wind and rain continues to be a maintenance problem, and periodically pile surfaces in eroded areas must be dressed and slack accumulations leveled, particularly at snow fences installed for control of slack deposition. As noted in the previous year, scattered and increasing weed growth on side slopes and top surface of the piles is an aid in the control of wind erosion.

Periodic observations are conducted on pile 5. Despite evidence of minor air infiltration at the surface, temperatures have remained at a safe low level, and no difficulty from spontaneous combustion has been encountered. Yearly checks of the heating value of pile depth samples, compared on a moisture- and ash-free basis, have shown a decrease of less than 1-1/4 percent from August 1953 to August 1957. During the year, in addition to the continuing observations, work included a determination of the size consist of a large channel sample removed from one of the piles and determination of the bulk density. Despite the extensive degradation caused by the compaction process, the size consist of the eight individual samples was remarkably uniform, and 23 percent of the combined sample was plus 2-inch and 72 percent was plus 1/4-inch. The average bulk density of the eight samples was 69.9 lb. per cu. ft.

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[35] Berger, L. B., Some Considerations on Dust Control in Coal Mining: AMA Arch. Ind. Health, vol. 15, No. 6, June 1957, pp. 499-505.

[36] Owings, C. W., and Harmon, J. P., Full-Shift Dust Exposure in Some Bituminous-Coal Mines: Bureau of Mines Rept. of Investigations 5361, 1957, 10 pp.

Freezeproofing Lignite

Producers supply lignite consumers with fuel as required, despite the severity of winter weather. Vaporization of moisture occurs during transport, and recrystallization as frost on coal particles at car walls and the exterior of cars cements particles together, frequently agglomerating the whole mass. Application of 3 to 5 quarts of oil per ton of lignite, as used by industry, is beneficial in reducing th degree of agglomeration, primarily by its influence on the structure of the frost deposited on the particles. In contrast to the dense, strong structure of crystals deposited on untreated particles of lignite those deposited on oil-treated particles are moldlike, delicate, fragile crystal structures and thus produce bonds of weak strength.

A method had been developed to determine the force necessary to rupture the agglomerated mass by measuring the pressure within a balloon imbedded in the center of the sample at the instant of rupture. Improved reproducibility of results has been obtained by equalizing the flow and turbulence throughout the refrigerated test chamber and by obtaining more uniform lignite samples. Testing and comparison of freezeproofing agents have been resumed.

Size Reduction of Lignite

In its natural state lignite is tough and resilient and resists fracture. Emphasis in the study of size reduction continues on a study of pulverization characteristics and power requirements.

Pulverization of lignite was continued in an experimental impact pulverizer loaned by the chemical engineering department of the University of North Dakota. Stable, controlled operation without using preheated air was achieved by removing an internal fan, designed to return larger particles to the grinding zone. External classification and return of larger particles to the grinding zone resulted in a product with high degree of fineness.

Mechanism of Flotation

A study is being made of the theory and practice of flotation of very fine sizes of coal that cannot be cleaned by gravity separation. The results of this study should lead to more efficient preparation methods, which in turn should contribute materially to extending our coal reserves.

During the year, continued emphasis has been placed on elucidating the mechanism of froth and kerosine flotation and determining the influence of operating variables on cell capacity. A comprehensive investigation of flotation rates and their relation to feed characteristics and operating variables has been undertaken, using flotation cells ranging in capacity from a few pounds to several tons per hour. Analysis of the flotation data indicates that the relation of the rate of flotation to the operating variables is complex, and a clear cut solution may not be possible, the order of the process depending on a number of variables.

Fine-Coal Cleaning

Present methods of cleaning fine coal are less effective and more costly than treatment of the coarser sizes. Improved methods could minimize the waste of fine coal now lost through inefficient cleaning, provide coal of better quality, and reduce costs.

One apparatus under investigation for several years (first in the laboratory and then on plant scale) is the feldspar jig, used widely in Europe to clean fine coal. The first installation of a modern feldspar jig in this country was made at a mine in Washington, where it has been subjected to a number of performance tests. These tests have not been completed and will be described in a forthcoming report of investigations.

In another attack on the fine-coal cleaning problem the dense-medium cyclone is being investigated at two laboratories (fig. 4). Three coals have been tested in a dense-medium cyclone washer; excellent cleaning efficiencies have resulted for two of them. Means have been studied for recovering medium from the cyclone products - the main problem that has prevented commercial adoption of this process for cleaning coal. Progress has been made in reducing magnetite consumption to an acceptable level.[37]

The viscosity of the dense medium used for cleaning coal plays an important role in cleaning efficiency.[38] Testing of mediums contaminated with coal and various types of clay demonstrated that the viscosity produced by a contaminant is related to its particle size. Bentonitic clays containing principally sodium montmorillonite, which disperse to extremely fine particles, increase viscosity the most. Kaolinitic clays do not break down to such fine size and influence viscosity much less. Identification of the principal minerals in a clay by routine differential thermal analysis permits a prediction of approximately how much it will affect viscosity. Cleaning trials with mediums of different viscosities and four coals of varied densities showed a steady reduction in cleaning efficiency as viscosity increased. However, the behavior of each coal was characteristically different.

Dewatering Coal Slurry

The growing number of preparation plants that must operate with closed water circuits to meet stream-pollution requirements focuses attention on the increasing need for research on liquid-solids separation. Investigations have been underway for several years, and a recent report shows how flocculation can

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[37] Yancey, H. F., and Geer, M. R., Laboratory Cleaning Trials on a Pennsylvania Anthracite in a Baum-Type Jig and a Dense-Medium Pilot Plant: Rept. of Investigations 5308, 1957, 23 pp.

[38] Geer, M. R., Sokaski, Michael, West, John M., and Yancey, H. F., The Role of Viscosity in Dense-Medium Coal Cleaning: Bureau of Mines Rept. of Investigations 5354, 1957, 64 pp.

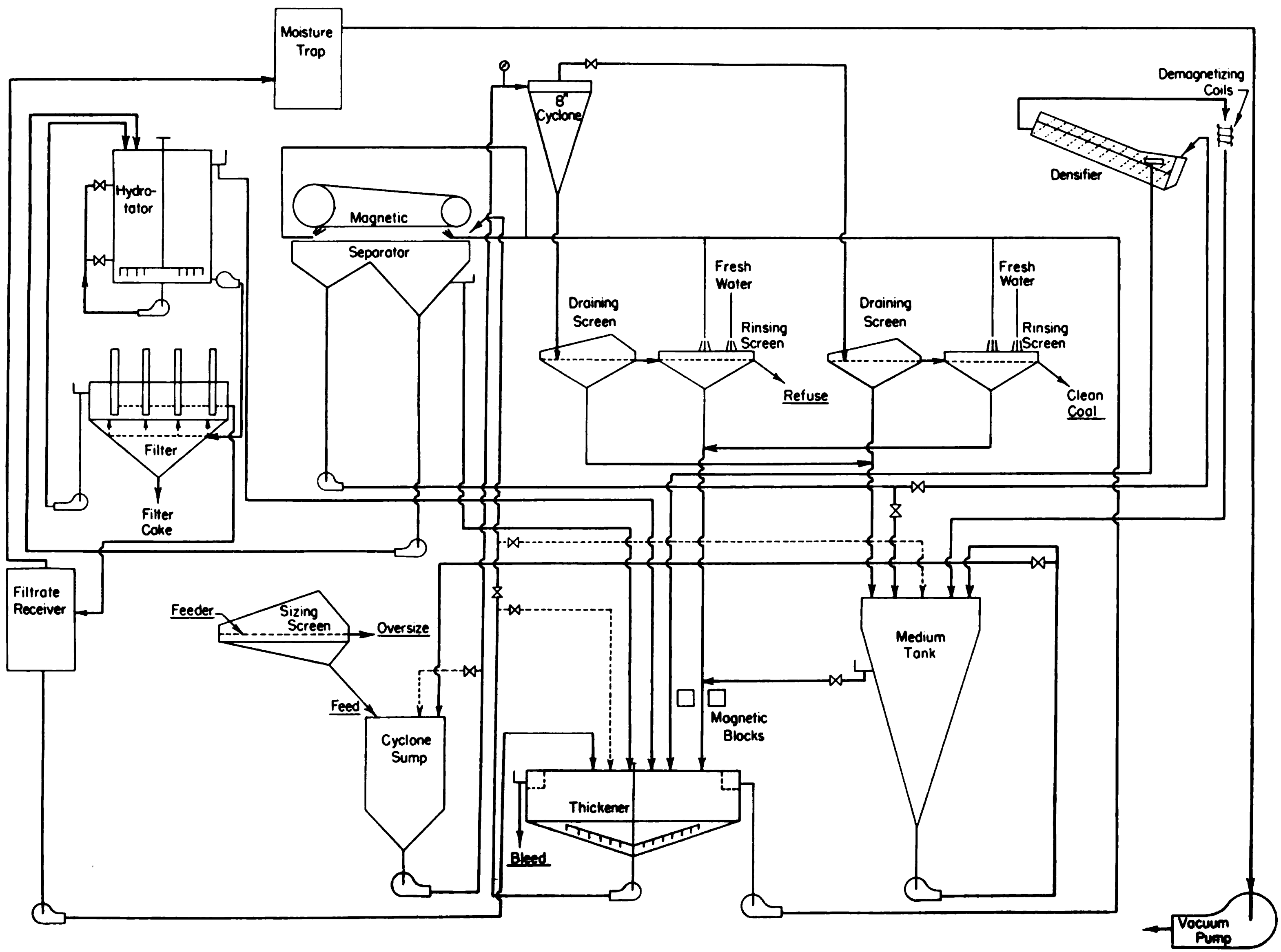

FIGURE 4. - Schematic Diagram of Coal-Preparation Plant With Dense-Medium Cyclone.

improve the vacuum filtration of some slurries.[39] A slurry that was virtually nonfilterable in natural form because it contained much fine material and clay was tested with 15 different flocculants in a test-leaf apparatus to determine the influence of flocculation on filtration. The flocculants varied widely in effectiveness, both as to the amount of increased filter capacity and as to the strength (and therefore ease of handling) of the flocs. With the better flocculants, cake rates that exceeded 200 pounds per square foot per hour were obtained, even on finely woven filter cloth.

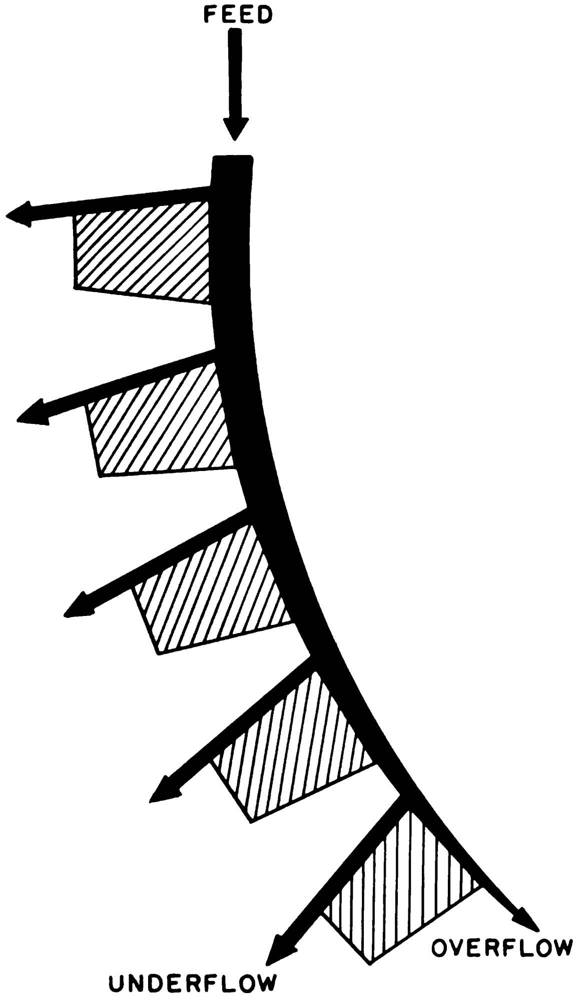

FIGURE 5. - Diagram of Sieve Bend for Cleaning Coal (After Fontein).

Closely related to the problem of dewatering slurry and equally difficult is the task of screening out fine sizes before further treatment. A radically new type of screen for this service is being tested. Developed in The Netherlands several years ago and known as the DSM (Dutch State Mines) screen or sieve bend, it is essentially a stationary section of wedge-wire screen bent to a 60° arc (fig. 5). As slurry flows down the screen, each successive wedge bar slices a layer off the bottom of the stream, carrying with it particles up to half the size of the net bar spacing. Thus a 1/2-mm. screen sizes at 1/4 mm. The laboratory tests have demonstrated that the screen has very high capacity and is capable of effective sizing in the difficult 1/4-mm. range.

Preparation Characteristics of Coking Coal

Coals are being tested for preparation characteristics on a county basis, initially in the Appalachian area. During 1957 reports for one West Virginia, one Kentucky, and one Virginia county were published. Figure 6 shows the 21 counties for which reports have been published and the status of the uncompleted counties. In addition, a New Mexico coal was examined for its washability characteristics.[40]

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[39] Jacobsen, P. S., and Mauser, J. E., Flocculation Improves Vacuum Filtration: Coal Age, vol. 62, No. 12, December 1957, pp. 74-75.

[40] Crentz, W. L., Washability Characteristics of Coal From San Juan County, N. Mex.: Bureau of Mines Rept. of Investigations 5335, 1957, 20 pp.

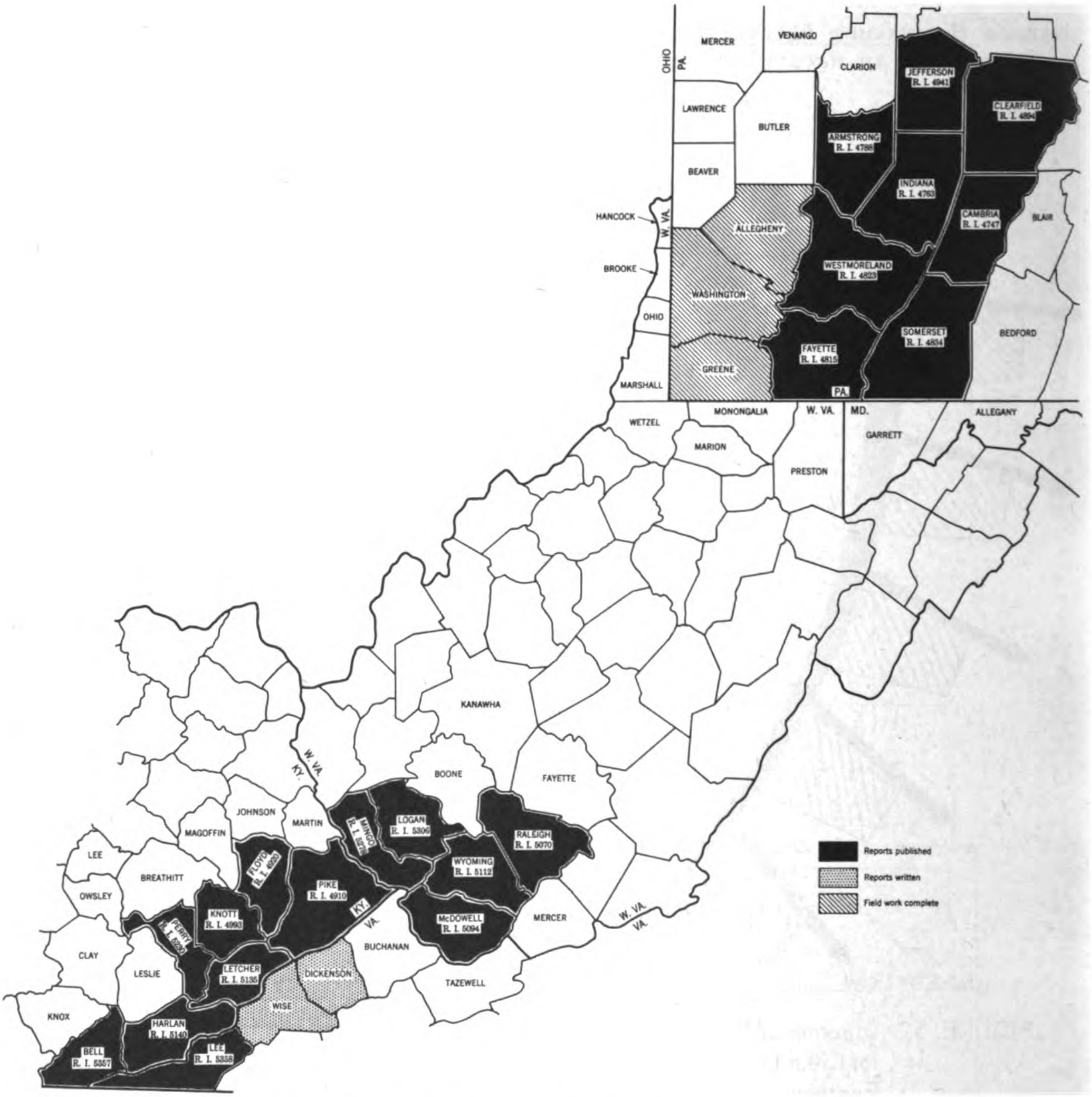

FIGURE 6. - Map Showing 1957 Status of Reports on Washability of Coking Coal.

Logan County, W. Va.

The Cedar Grove group and the No. 2 Gas coalbeds are important from both the reserve and production standpoints and can be prepared by mechanical cleaning to yield coal suitable for metallurgical use.[41] The Cedar Grove group of coalbeds consists of four mining horizons; each horizon is treated as a separate bed to report the preparation characteristics effectively, except that the Upper Cedar Grove bed could not be sampled and studied. However, all coal produced from the Cedar Grove group was combined in the table of production. The Chilton bed also is important from a production standpoint, and its coal can be prepared to produce a chemically suitable metallurgical fuel. The Alma bed has important reserves in the county but is unimportant in production. In some areas this coal can be upgraded to metallurgical fuel and in others it cannot because of a variable sulfur content. The No. 5 Block, Stockton, and Buffalo Creek coalbeds have minor importance from both production and reserve standpoints, but the coal from these beds may be prepared to produce a low-ash product chemically suited for metallurgical use.

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[41] Miller, J. W., and Jolley, T. R., Preparation Characteristics of Coal From Logan County, W. Va.: Bureau of Mines Rept. of Investigations 5306, 1957, 48 pp.

Logan County coals generally are not responsive to fine crushing as a means of product improvement. The total production of coal in Logan County in 1953 was 17,190,007 tons, of which approximately 68 percent was cleaned in 24 mechanical cleaning plants. The Cedar Grove group, the Chilton, and the No. 2 Gas coalbeds were the sources of most of the cleaned coal.

Bell County, Ky.

The Dean, Mingo, and Harlan beds are the major sources of production for the county, and the Mingo, Poplar Lick, and Jellico coalbeds have the largest reserves.[42] Two faults divide the county into three coal-bearing areas - Log Mountain, Puckett Creek and Hance, and the area north of Pine Mountain, in which the correlation of beds is difficult. The preparation characteristics of coal in each of these areas were determined.

In the Log Mountain area coal from the Red Spring, Hignite, Stray, and Mingo beds can be prepared to produce fuel for metallurgical use. The Sterling coalbed cannot be considered a source of metallurgical fuel under present standards because of high sulfur content. Coal from the Sandstone Parting bed can be washed mechanically to yield a metallurgical fuel.

In the Puckett Creek and Hance area coals from the Creech and Harlan beds can be prepared to yield a chemically suitable product of Metallurgical grade. The Imboden coal in the western part of the area can be prepared to produce a low-ash, medium low-sulfur product suitable for making metallurgical coke; in the eastern part of the area the sulfur content of this coal is too high under present standards for coking coals. The Dean coalbed varies in sulfur content, and for this reason Metallurgical-grade coal may be prepared from some areas but not from others.

In the area north of Pine Mountain the Moss bed cannot be considered a source of metallurgical fuel because of the difficulty of reducing the sulfur content by present cleaning methods. Coal from the Jellico bed can be prepared by an easy washing operation to produce a low-ash, low-sulfur product chemically suited for metallurgical use.

42/ Jolley, T. R., and Miller, J. W., Preparation Characteristics of Coal From Bell County, Ky.: Bureau of Mines Rept. of Investigations 5357, 1957, 30 pp.

In 1954 the total production of coal in Bell County was 1,247,160 tons, of which 30.7 percent was cleaned in five mechanical cleaning plants. Most of the coal cleaned came from the Mingo and Dean beds. Little benefit would be gained by finer crushing for beneficiation, because of the difficulty in handling the excess fines in a commercial cleaning plant.

Lee County, Va.

Only a small area in the north central part of the county supports mining.[43] Beds in this area are difficult to correlate because of geologic movement. The Taggart coal bed is the major source of production, and the Kirk, Harlan, Imboden, and Pardee coalbeds have minor importance. Total production was 660,395 tons in 1955, of which 38,756 tons from the Taggart bed was cleaned on air tables.

Coal from the Taggart and Imboden beds is suited for metallurgical use as mined. Coal from the Kirk and Harlan beds cannot be upgraded to metallurgical use under present standards because of excessive sulfur content. Lee County coals do not generally respond to finer crushing for product improvement.

Pierce County, Wash.

In addition to the regular coking-coal survey, which is confined to the Appalachian region, washability examinations were made of samples from a deposit in Pierce County, Wash., which contains the only known reserves of coking coal on the Pacific coast. This work was undertaken in cooperation with a company that is exploring the deposit. The coals examined have high free-swelling indexes and low sulfur contents but are difficult to clean and cannot be reduced readily to less than 10 to 11 percent ash.

Anthracite Preparation

Activities in the preparation field have been confined largely to the establishment of a laboratory suitably equipped for the study of special preparational procedures; the objective is to provide quality anthracite products for new demands of the changing market.

Heyl-Patterson cyclones have been installed to study desliming, sizing, and deashing of fine sizes of anthracite and to supplement equipment used for studying reduction of the ash content of fine-size, heavy-medium feed. A drum-type, heavy-medium pilot plant has been installed, and enough tests were completed to determine the operating characteristics of the unit. A unitized, air-swept, ball-mill system has also been installed to study size reduction of anthracites along with the effect of size on various cleaning processes. A 10-compartment, washability-assay tank has been installed in the preparation laboratory, and separations are possible at 1.40 to 2.00 specific gravity at 0.05 increments. Tests conducted on preparation of minimum ash fractions from conventional domestic coal sizes have indicated that for a low-volatile, hard-structure coal increments containing as little as 1.25 percent ash are possible.

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[43] Miller, J. W., Preparation Characteristics of Coal from Lee County, Va.: Bureau of Mines Rept. of Investigations 5358, 1957, 14 pp.

For a high-volatile anthracite studied, increments containing as little as 0.75 percent ash have been prepared. A sample of mine-run anthracite has been obtained for study in the heavy-medium plant, and samples from conventional breaker operations have been received for study in the cyclone system.

A study of the preparation required for anthracite feed to a Lurgi-type gas-producer plant has been initiated in conjunction with the gasification investigation. This study covers two possible preparation procedures that yield products suitable either for operation of a gas plant in conjunction with a large, modern, conventional mining and preparation operation or for a new mining operation wherein the entire output from the mine would be used in a large, commercial gas plant. The first type of operation entails collecting all plus-3/16-inch refuse, crushing to 9/16- by 3/16-inch, and separating to produce a gas-plant feed with 20- to 25-percent ash content. This type of operation, however, limits the size of the gas plant but makes possible a low-cost fuel. The second type of operation places no restriction on the size of the plant but does require anthracite production at a price lower than is possible with mining procedures as now developed. If the cost of mining should be reduced appreciably, this type of operation will offer a far more versatile type of gasification operation.

COMBUSTION, BOILER-WATER TREATMENT, AND POLLUTION

Heat Transfer in Furnaces

Efficient use of fuels for producing heat and power is of major importance in our industrial economy.[44] A continuing program in this field has included studies of heat transfer in steam-boiler furnaces and of ash and slag deposits that form on the external surfaces of boiler tubes, where they reduce heat flow and hinder gas flow through the furnace.

Field tests and analyses of data have been completed in the sixth of a series of tests on large steam-boiler furnaces, carried out in cooperation with the American Society of Mechanical Engineers. Theoretical flame temperatures, based on methods of computation previously described, have proved useful in correlating the performance of the different furnaces. These correlations have been based upon the theoretical flame temperature as a measure of the initial heat-transfer potential and the residence time of a given volume of gas in the furnace.

Deposits on the external surfaces of boiler tubes of ash and slag that bear little resemblance in composition or physical form to the mineral content of the coal fired have been a subject of continued study. The mineral content is released from the coal as particles of varying size, composition, and fusion temperature. Depending upon local conditions of temperature, gas composition, and agglomeration or lack of agglomeration, ash may leave the combustion zone as molten slag, fly ash, or volatilized matter. The type of deposit that forms on a given surface depends upon the particular ash components conveyed to that surface and upon the conditions of temperature and gas composition. Experiments are being continued in synthetic furnace atmospheres on changes in composition and physical state of fly ash subjected to high temperatures and high-temperature gradients. Chemical and X-ray analyses show migration of ash components within the deposits. The release of ash components from coal will be considered in relation to the study of pulverized coal flames.

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[44] Corey, R. C., Discussion of An Effort to Use a Laboratory Test as an Index of Combustion Performance, by F. J. Ceely and R. I. Wheater: Trans. ASME, vol. 79, No. 5, July 1957, pp. 1177-1184.

Results were published of a study of burning solid fuels in thin beds.[45]

Combustion of Coal in Suspension

An increasingly large fraction of coal used in large steam-boiler furnaces is burned as pulverized coal in suspension. Equipment is being developed for studying pulverized-coal flames burning 1 to 5 pounds of coal per hour. Flame velocities and other conditions needed for producing well-defined flames are known for small flames from a few milligrams per minute to a few grams of coal per hour. One purpose of the investigation is to extend such measurement to higher burning rates, and another is to determine the effect of local intensity of combustion upon the release of ash and subsequent formation of ash deposits.

Fluid Mechanics

Fluid flow patterns, turbulence, and momentum interchange often control and always significantly influence combustion reactions. A survey will be made of developments in this field that are applicable to the study of combustion equipment. Also, a background of experience in fluid mechanics will be obtained for application to other projects.

Activity during 1957 included reviews of a new book[46] and of work in this field published the preceding year.[47] Also included were field studies of hydraulic transport of coal from mine to washing plant. Transport with a centrifugal pump through a 6-inch line was feasible, but size degradation was excessive, probably due in large part to the pump. Further work will be needed to determine the effect of different pump designs.

Combustion of Anthracite in Small Industrial Stokers

Because virtually no information is available on the characteristics of industrial stokers in the 100- to 400-pound-per-hour class, technologists have conducted tests on four stokers, classified as small, industrial anthracite burners. Preliminary runs were made on a Coxe traveling-grate stoker and on a Skelly underfeed, side-retort, moving-grate bar stoker (fig. 7). The maximum heat efficiencies obtained in these trial runs were 63 percent with the Coxe and 70 percent with the Skelly stoker. The chief difficulties experienced in these operations were: (1) The average CO_2 content of the flue gases for any run did not exceed 9 percent, probably the result of excessive air flow; and (2) the varying combustible content remaining in the ash-pit refuse was high - 20 percent in the two best runs on the Skelly stoker, 37 and 43 percent in the Coxe stoker. The Skelly stoker was thoroughly reconditioned before another series of tests was made. In these tests the maximum hearth efficiency was 78 percent; CO_2 in the flue gases ranged from 9 to 12.5 percent; and combustibles in the ashes ranged from 23 to 75 percent.

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[45] Carman, E. P., Graf, E. G., and Corey, R. C., Combustion of Solid Fuels in Thin Beds: Bureau of Mines Bull. 563, 1957, 92 pp.

[46] Weintraub, Murray, Book Review of Momentum Transfer in Fluids by Corcoran, Opfell, and Sage: Ind. Eng. Chem., vol. 49, No. 5, May 1957, pp. 83A-84A.

[47] Weintraub, Murray, Flow of Fluids: Ind. Eng. Chem., vol. 49, No. 3, March 1957, part II, pp. 497-502.

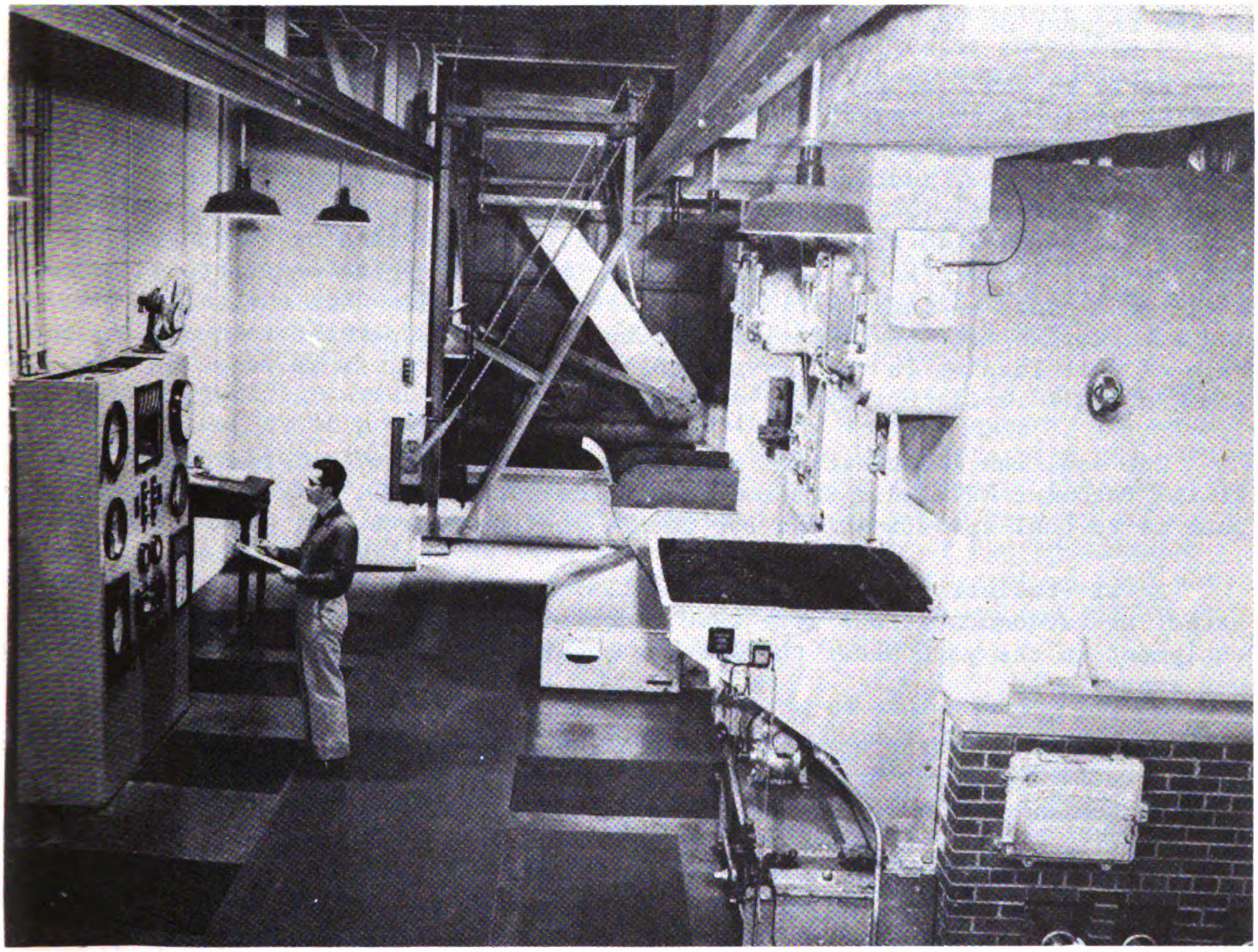

FIGURE 7. - General View of Boilerroom Showing, Two Stoker-Boiler Combinations, Instrument Panel, Coal Hopper, and Larry Car.

The stoker study included tests of two water-cooled, reciprocating-pan, crossfeed stokers installed successively in place on the Skelly stoker. On these new units (which were essentially the same size, construction, and operation, although manufactured under different trade names) 31 runs were made at fuel rates ranging from 63 to 287 pounds of wet coal per hour. The CO_2 contents averaged 10.8 to 11.5 percent in the two tests, and heat efficiencies ranged from 69 to 84 percent; there was very little difference between the two units. Peak efficiencies of 83 and 82 percent were obtained at about 80 percent of the boiler manufacturer's rated output (100 percent equals 2.5 million B.t.u. per hour). The carbon content of the ashpit refuse ranged from 12 to 43 percent. The average carbon content of the ashes for the two tests was 22 and 25 percent.

Fuel-Engineering Service

Engineering assistance and technical advice were provided to Federal agencies in selecting, testing, and using fuel and fuel-burning equipment. At the request of the agencies listed, the following services were rendered:

Early in the year tests were run on the new 150,000-lb.-per-hr. power and heating boiler at the Naval Gun Factory, Washington, D. C. A low-volatile and a high-volatile coal were used to determine whether this equipment could burn these coals at low, medium, and high rating without exceeding limitations on smoke and dust emission. With both coals these limitations were not exceeded at the low and medium rates of operation but were exceeded by a wide margin at full rating. After the equipment had been modified by the manufacturer, complete continuous-rating, overload, and efficiency tests were run. The boiler and accessories met specifications, except for high superheat temperatures and rate of dust loading in the fly-ash collection hoppers; smoke emission from the stack at full rating barely met specifications.

The coal was inspected and operation was observed of the heating boilers at the Patuxent Naval Air Station, Lexington Corners, Md. Recommendations were made as to coal specifications for future coal purchases.

An air-pollution problem in the Philadelphia Naval Shipyard was investigated, and recommendations were made concerning operation of the offending equipment in the yard under the weather conditions prevailing when most trouble was experienced.

Problems regarding corrosion of the gas side of air preheaters for boilers at the Naval Shipyard, Bremerton, Wash., were discussed with the staff there, and recommendations were made for alleviating the difficulties.

Operating conditions, damaged grates, and coals being used at the New York Naval Shipyard, Brooklyn, N. Y., were examined and discussed with the staff there, and recommendations were made to segregate coals when shipments from more than one contractor were received and to burn the coals separately. Later, the performances of different coals in the boilers were observed, and recommendations were made to improve fuel-bed conditions.

Boiler-Water Treatment

Condensate Corrosion

Federal agencies spend millions of dollars annually for maintenance work involving the replacing of valves and piping in the large condensate-return systems that save water, fuel, and chemicals by returning the condensed steam for boiler feedwater. This waste stems from the impurities that are brought into the boiler with the raw makeup water. It is cumulative because larger condensate losses from leaky return lines increase the raw-water makeup requirements and corrosion therefore occurs more rapidly. Boiler corrosion from dissolved oxygen in the feedwater also is promoted when condensate-return lines fail, because feedwater heaters are not often sized to deaerate unusually large quantities of cold makeup.

A consendate-corrosion tester was developed to survey corrosion problems. The tester is essentially a nipple in which the average rate of pipe-wall thinning is obtainable from weight loss and the factors responsible for the corrosion are disclosed by the corrosion pattern. The tester therefore provides information both on the overall corrosivity of the steam condensate and on the extent to which corrosion tends to be localized in the form of channeling or pitting. A total of 465 such testers were evaluated during the year, and 328 new tests were begun.

When a survey discloses a corrosion problem, operating practices at the plant are reviewed as the first step toward control. Problems have been solved by decreasing the carbonate content of the feedwater, better deaeration, use of cation-exchange resins, distillation, or chemical treatment.

The principal current use of the Bureau of Mines tester is to determine the effectiveness of preventive chemical treatment or the suggested improvements in operating practices. A considerable number of the testers have shown that decreasing the carbonate content of the feedwater decreases the severity of the corrosion problem. Neutralizing amines were found to be very effective when the dosage was sufficient to neutralize the acidity of the condensate. However, the tester results have also shown that the filming amines have not been consistently satisfactory because the nipples were wormed and pitted at nearly half of the plants where filming treatment was used.

The boiler tests showed that an apparent loss of amine during steam distillation of amine recovery in the condensate is a reasonable criterion. Because no amine was found in the blowdown, the only logical conclusion is that part of the amine was degraded to something that did not react like octadecylamine with bromphenol blue or the other analytical agents. This disappearance was traced to the probable coalescence of octadecylamine to dioctadecylamine, a secondary amine.

Indications are that dimerization to dioctadecylamine occurs both in hot water and in steam. However, most of the secondary amine formation was shown to occur during condensation of steam containing octadecylamine. Therefore, it should make little difference with respect to degradation whether the amine is fed to the boiler for steam distillation or pumped into the export steam.

Octadecylamine alone, when pumped continuously into the condensate, developed a good corrosion-inhibiting film on the tester metal. However, contamination with sodium sulfate resulted in a decrease in film continuity, so that the tester surfaces were pitted. Condensate containing sulfate and octadecylamine caused deeper corrosion than condensate to which neither had been added.

To test laboratory findings in an operating plant, provision was made for sampling at various points in a Department of the Army heating system that is given octadecylamine treatment. Approximately 50 pounds of amine was pumped into the steam over a period of several months. At no time has octadecylamine or dioctadecylamine been found in the condensate returned to the boiler. However, an unusual increase to more than 2 p.p.m. was noted in the ammonia content of the condensate. This ammonia could not possibly be attributed to the makeup water monitored. Much of the amine appears to collect in steam traps and a large percentage of it is in the form of the secondary amine.

Engineering Service

Engineering assistance was given to Federal agencies on water problems. In some instances considerable laboratory work was involved. Special assistance to the General Services Administration included the assay of samples submitted with bids on Federal stock items for boiler-feedwater treatment. Also, the final draft of an interagency-approved Federal specification for sodium polyphosphate was reviewed for the accuracy of its technical content. Assistance of a similar nature was given to the Cataloguing Division on its listing for quebracho tannin. The Department of the Army was assisted with a variety of water problems, including overall policy on water supply. Special assistance was given in several instances on problems of corrosion in potable-water piping. Other problems involved corrosion and deposition in cooling systems and corrosion in hot-water piping. The Department of Agriculture was assisted with a cooling-water problem, and the Capitol architect was helped with the specifications and operation of the new Capitol air-conditioning system. The Veterans Administration was given special assistance with several contamination problems which had led to deep pitting of hot-water piping. Assistance to the Department of the Air Force included evaluation of the test specimens from two embrittlement-detector surveys. There were also the usual problems of corrosion in hot-water piping and in refrigeration systems. Special assistance to the Department of the Navy included the solution of silica-contamination problems at two plants that generate electricity. A third generating plant had trouble from deposited iron oxide. Special advice was given on a formal program of corrosion testing at the Naval Boiler and Turbine Laboratory at Philadelphia. Examination was made of ion-exchange resins, the exchange capacities of which had deteriorated. The Navy also had some peculiar scale and sludge problems as well as the usual problems of corrosion and deposition of corrosion products.

Work continued on the analysis of boiler water and miscellaneous raw and return samples from Government agencies, and the analysis resulted in recommendations on boiler water treatment. During the year a total of 14,054 samples of water were analyzed for these agencies and recommendations were made. Reports and recommendations were also made covering analyses of 6 boiler-plant scales and deposits, 292 miscellaneous special water samples, and 3 boiler-water-treatment compounds. More than 3,000 orders from Government agencies were distributed for use by engineers for testing boiler water.

Consulting service was provided many agencies on boiler-water treatment, prevention of corrosion, scale, and sludge deposits, prevention of contamination by oil, and related problems. The number of plants being serviced increased about 2 percent during the year, bringing the total number of boilers serviced to an estimated 9800. Through operation of the program substantial savings are being realized by Government agencies in increased boiler efficiency and lower plant maintenance and repair costs. Some representative plants report savings of $3,400 per boiler per year in fuel, maintenance, and replacement costs.

Air Pollution

Combustion Characteristics of Refuse

Investigation of combustion characteristics of refuse has continued. The objective of this work is to obtain information for designing and operating incinerators capable of producing completely innocuous discharges.

Conditions necessary for ignition, rates of burning, and rates of penetration of thermal and combustion waves into the refuse bed have been determined in a cylindrical furnace with 100 percent overfire air fed from tangential ports. The refuse, used as fuel, was a blended mixture of wood chips, shredded cardboard, paper, and enough shredded lettuce to give a moisture content of either 25 or 50 percent. The burning rate increased with air-flow rate, decreased with increased moisture content, and was not affected by the hood temperature provided it was adequate for ignition, at least 1,600° F. The rates of penetration of the evaporation and combustion waves depended upon moisture content, air rate, and level in the bed, with a possible effect of the hood refractory temperature on spacing of the separate waves.

Disposal of Radioactive Wastes

Cooperative work with AEC continued on incineration of radioactive wastes during 1957. A paper reporting results of this study has appeared in the Fuel Society Journal of the University of Sheffield; other publications are planned.48/

Air Pollution by Sulfur Dioxide

In cooperation with the Public Health Service, the Bureau is investigating methods for decreasing air pollution resulting from sulfur dioxide in flue gases and unburned hydrocarbons in automotive exhaust. Most of this sulfur dioxide is evolved in the air of the larger cities. Sulfur dioxide is highly irritating to man's respiratory system, deleterious to plant life, and injurious to metals, fabrics, and building materials.

48/ Corey, R. C., and Schwartz, D. H., Combustion of Low-Ash, High-Volatile Solids With Tangential Overfire Air: Fuel Society Jour., University of Sheffield, vol. 8, 1957, pp. 5-13.

In previously developed processes for absorption of sulfur dioxide, absorption in aqueous solutions has been used almost exclusively. Cost estimates have been prepared for three of these processes.49 50/

The experimental program has been directed toward developing cyclic absorption processes for removing sulfur dioxide. Bench-scale investigations include: (1) Absorption by solid oxides of copper, manganese, lead, calcium, iron, and aluminum; (2) adsorption with impregnated charcoals; and (3) regeneration of absorbents.

Temperatures available for absorption are either in the range 250° to 300° F. (at which the flue gas leaves the air preheater before entering the stack) or 600° to 650° F. (the temperature of the flue gas before it enters the preheater). Absorption in the higher range now under investigation offers certain advantages: (1) The rates of reaction are considerably higher, greatly extending the field of potential absorbents; and (2) removel of sulfur dioxide would eliminate the corrosion problem now encountered in the air preheater of powerplants. Methods of regenerating spent absorbent and recovering sulfur products under study are: (1) Thermal decomposition of the sulfide or sulfate; (2) reduction of spent absorbent with a reducing gas such as hydrogen or methane; and (3) electrolysis of the metal sulfate to produce sulfuric acid.

Air Pollution by Automobile Exhausts

Air pollution from the exhaust gases of an automobile engine might be avoided by catalytic oxidation. A complicating factor is poisoning of the catalyst by lead deposits from leaded gasoline. Thus, in the initial program, development of effective catalysts in the absence of lead compounds and tolerance of lead are being studied. One-component catalysts - oxides of chromium, iron, nickel, copper, and aluminum - have been investigated between 200° and 600° C. with several hydrocarbons. Tests to determine the amount and nature of lead deposits have just begun.

Stream Pollution

Little work was done on this subject during 1957, owing to lack of personnel. At the invitation of the American Institute of Engineers a paper was written on the utilization and disposal of waterborne industrial wastes.51/

49/ Field, J. H., Brunn, L. W., Haynes, W. P., and Benson, H. E., Cost Estimates of Liquid-Scrubbing Processes for Removing of Sulfur Dioxide From Flue Gases: Jour. APCA, vol. 7, No. 2, August 1957, pp. 109-115.

50/ Field, J. H., Brunn, L. M., Haynes, W. P., and Benson, H. E., The Costs of Scrubbing Out SO_2 From Flue Gases: Combustion, vol. 29, No. 5, November 1957, pp. 61-66.

51/ Berk, A. A., Utilizing and Disposing of Waterborne Industrial Wastes: Min. Eng., vol. 9, No. 7, July 1957, pp. 780-783.

CARBONIZATION AND ITS PRODUCTS

Thermal Decomposition of Coal

Knowledge of the mechanism and kinetics of tar formation and its conversion at higher temperatures to high-temperature tar is important in the carbonization industry. The mechanism of tar formation appears to be intimately associated with the mechanism of coke formation, development of coke strength, and expansion characteristics of the coal; thus, knowledge of tar formation and its reactions will lead to better understanding of the coking mechanism.

As one means of studying the mechanism of coke and tar formation, the Bureau is studying the composition and yields of gases, liquids, and residues formed by carbonizing small samples of coal under various pressures at 300° to 550° C. At constant carbonizing temperature the total volume of gas decreased as the absolute pressure above the coal decreased.

In a related study a bed of coal fluidized by steam or nitrogen has been used, and provision was made for recovering the overhead products directly, as in conventional carbonization practice or after passing them through a cracking unit at 600° to 800° C. (fig. 8). Superficial velocities of the fluidizing mediums of 3.5 to 3.9 feet per second are best for tar production when a mixture of 14- by 35-mesh high-splint coal and char is fed. Char yields from the fluidized carbonizer are much lower than those obtained in the BM-AGA apparatus; but tar yields are much greater, and the tar contains much more pitch. Fluidization with steam or nitrogen produced essentially the same yields of acids, bases, aromatics, olefins, and paraffins. Except for approximately equal yields of olefins, yields of these products from fluidized carbonization are less than those from BM-AGA carbonization. A preliminary study in a thermal cracker shows that, when the cracking temperature is increased from 600° to 700° to 800° C., the yield both of the liquid portion of the tar (boiling below 350° C.) and of the pitch decreases progressively. This same behavior is observed in BM-AGA carbonization.

Expansion During Carbonization

Expansion test data are of particular interest to coke-oven operators because of the danger of oven damage from coals or blends that expand excessively during coking. As part of the survey of carbonizing properties of American coals, expansion tests in the sole-heated oven are made only on those coals that contain over 65 percent dry, mineral-matter-free fixed carbon.[52] Experience has shown that most coals of lower rank contract during carbonization. During 1957, 14 coals from Pennsylvania and West Virginia were tested. Although the sole-heated oven requires a charge of 18 kilograms of coal for a test, a much smaller-scale test with only 71 grams gives results that correlate well with the larger scale test and may well be useful in carbonization studies, particularly where the amount of sample is small.

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[52] Naugle, B. W., Wilson, J. E., and Smith, F. W., Expansion of Coal in the Sole-Heated Oven: Quantitative Effects of Dry Bulk Density, Moisture Content, and Particle Size: Bureau of Mines Rept. of Investigations 5295, 1957, 15 pp.

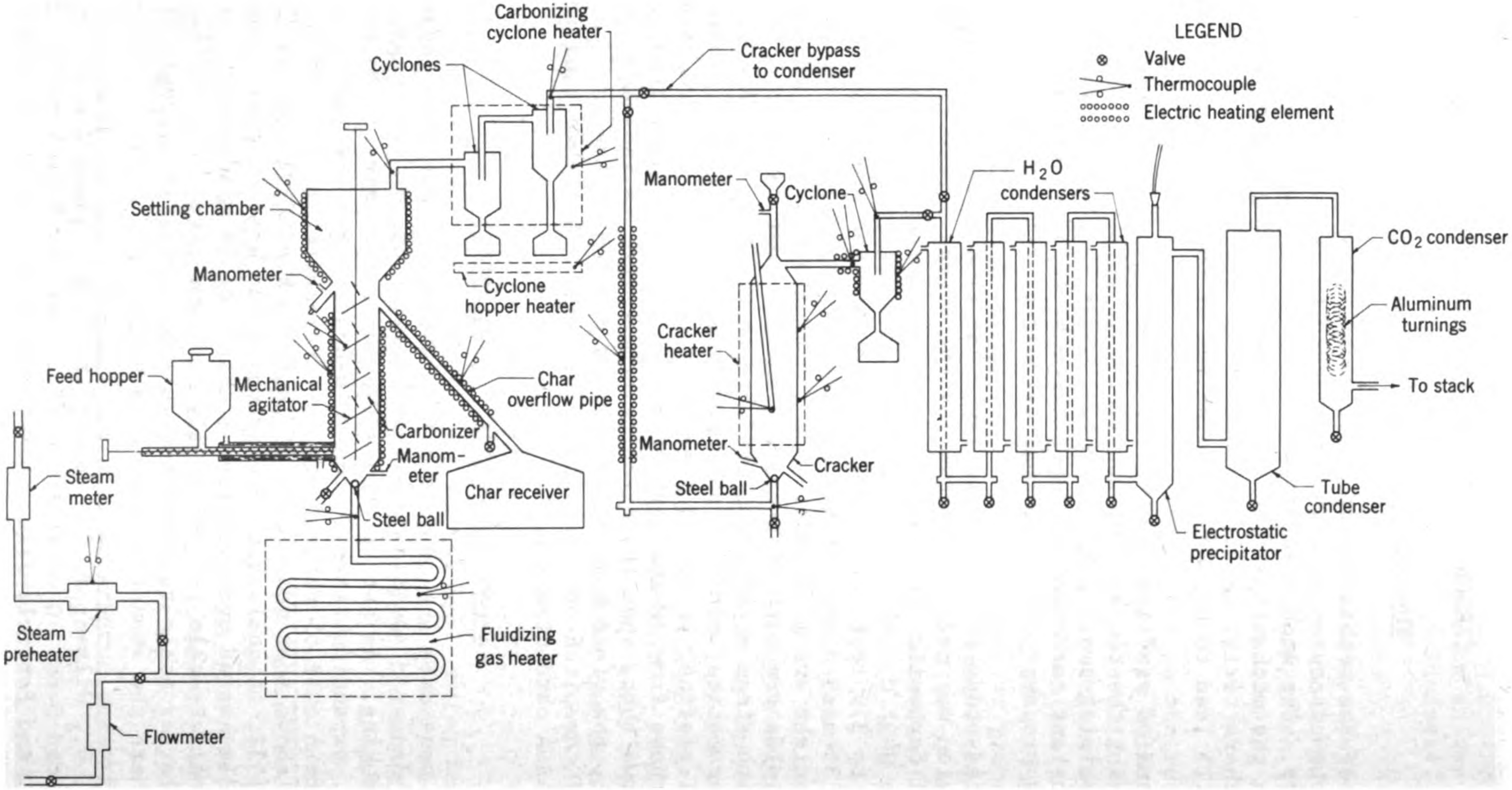

FIGURE 8. - Equipment for Studying Mechanism of Coal Carbonization.

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Effect of Pretreatment on Carbonization

Pretreatment of low-rank coals, particularly at temperatures below their softening points, improves the quality of the cokes obtained from their subsequent carbonization. This finding has special significance concerning production of improved metallurgical coke from western coals.

A high-volatile western coal, pretreated in an inert atmosphere at 215° C. and subsequently charged hot to a test oven, yielded a coke with a tumbler stability index twice that of coke obtained from direct carbonization. Pretreatment was less effective when the hot charge was allowed to cool before final carbonization. In another study a blend of Hiawatha coal from Carbon County, Utah, with 15 and 25 percent of a medium-volatile mixture of three coals from the Wilkeson area, Wash., yielded a stronger coke when preheated to 320° C. in steam than with no pretreatment, the tumbler stability indexes being 37 compared with 28, and the tumbler hardness indexes being 63 compared with 59.

Effect of Cleaning on Carbonization

A program on the effect of cleaning on coal carbonization was initiated in 1957. The mineral matter from which coal ash is formed may be considered a naturally occurring inert material in coal that affects the structure of coke, as do other inerts that are sometimes intentionally added to coal before carbonization. The petrographic composition of coal and hence the structure of its coke may also be altered by cleaning. Thus, removal of fine coal almost certainly results in removal of most of the fusain, small amounts of which result in a blockier coke.

Stockton-Lewiston coal from Kanawha County, W. Va., was cleaned at 1.45 specific gravity. The following samples were carbonized in the BM-AGA 18-inch retort at 900° C.: (1) Unwashed coal, (2) float fraction of the washed coal, and (3) float-and-sink coal recombined in proportions equivalent to the unwashed coal. The principal effect of cleaning was a reduction in ash content from 18.7 to 9.2 percent. The sulfur content of the unwashed coal was low (0.6 percent), and cleaning had no effect on it. Washed coal yielded coke with a lower 1-1/2-inch shatter index but higher tumbler indexes. This effect accords with previous data, which had shown that the addition of inerts increases the shatter index but decreases the tumbler indexes. The recombined sample of cleaned coal and refuse showed the same carbonizing properties as the original coal.

Effect of Fluidity on Abradability of Coke

The fluidity of a coal, as measured by the Gieseler plastometer or a similar instrument, is commonly believed to be a decisive factor in determining the strength of coke made from it. Fluidity may be changed by additions of inert material in fine sizes, such as high-temperature coke, silicon carbide, or activated carbon; the strength of the char prepared from the mixture is measured by a modified Roga abrasion test.

Two coals, one from the Lower Freeport bed of Clearfield County, Pa., having a maximum Gieseler fluidity of 15,000 dial divisions per minute, and the other from the Upper Freeport bed of Cambria County, Pa., having a maximum Gieseler fluidity of only 44 dial divisions per minute, were blended with varying proportions of an inert. Much larger proportions of inert were necessary to reduce to a minimum the strength (abrasion index) of the char obtained by carbonization of the coal-coke blends containing the more fluid coal. The strength of the char increased rapidly in the fluidity range of zero to five dial divisions per minute. Beyond this range the char strength was virtually independent of the fluidity. The difference in the char strength for the two coals of different levels of maximum fluidity was small when the fluidities of the respective blends were five dial divisions per minute.

The specific character of the inert material greatly affected the strength of char made from binary blends with coal. Silicon carbide appeared to narrow the plastic range of a coal much less than activated carbon when increasing amounts were added to progressively decrease the plastic range of this coal. Greater porosity and adsorptive capacity of the activated carbon limit the amount of cementing material available for binding the particles together. The abrasion indexes of chars obtained from blends containing small proportions of either silicon carbide or activated carbon and large proportions of coal were slightly higher than that of char from the unblended coal.

Coking Properties of Western Coals

In the cooperative program on improving the quality of coke made from western coals 21 high-temperature carbonization tests were run in the 500-pound, 14-inch, slot-type coke oven. Auxiliary studies were also made in the 50-pound, 10-inch modified BM-AGA retort, the 100-gram carbonization test, and the Parry-Potter dilatometer. Results and discussion of the complete series of 30 slot-oven tests and all auxiliary studies have been reported to the cooperating organizations. In general, the results show that the average size of coke decreased when the carbonization temperature was increased. The plus-1.06-inch tumbler index decreased slightly when the carbonization temperature was increased. Increased carbonization time in the experimental slot oven was shown to be proportional to the increased bulk density. Coke produced at low bulk densities generally had better physical properties than corresponding cokes produced at higher bulk densities.

Carbonizing Properties of Bituminous Coals

The objective of the continuing survey of carbonizing properties of American coals, in the BM-AGA apparatus to determine yields and properties of coke and other products at a carbonizing temperature of 900° C., is to evaluate the carbonizing properties of the principal coalbeds of the United States.[53] This evaluation will provide the metallurgical industry with information about the suitability of coals for making metallurgical coke. During 1957, 22 coals from West Virginia and 4 from Pennsylvania were tested.

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[53] Smith, F. W., Reynolds, D. A., and Wolfson, D. E., Coking Properties of Pittsburgh District Coals: Trans. AIME, Min. Eng., vol. 9, No. 3, pp. 360-364.

Boone County, W. Va.

Eleven coals, representing the Stockton-Lewiston, Coalburg, Hernshaw, Chilton, Cedar Grove, and Alma seams, ranked as high-volatile A bituminous. The ash content of nine of the coals ranged from 3.3 to 7.8 percent. One sample of Stockton-Lewiston coal contained 12.0 percent ash, and a sample of Alma coal contained 10.0 percent ash. These coals yielded rather abradable coke, which would probably be improved by cleaning to reduce the ash. Other samples of coal that were obtained from these beds, but contained less ash, yielded a less abradable coke. The two samples of Alma coal were high in sulfur, containing 2.6 and 2.7 percent; this factor would detract from their usefulness as a source of metallurgical coals under present standards. The other coals from this county would be satisfactory as a major constituent of coking blends.

Nicholas County, W. Va.

Five coals, representing the Lower Kittanning, Middle Kittanning, Eagle, and Sewell seams, were low in ash and sulfur content, which ranged from 3.2 to 6.8 and 0.7 to 0.8 percent, respectively. All the coals except the Sewell, which was lowest stratigraphically, ranked as high-volatile A bituminous. Sewell coal ranked low in the medium-volatile classification. The coals yielded coke having satisfactory physical properties for metallurgical coke with tumbler-stability indexes ranging from 38 to 57 and tumbler hardness from 61 to 66.

Logan County, W. Va.

Coals from the Alma and Eagle beds were high-volatile A in rank and had satisfactory chemical properties for metallurgical coals. Their coke properties indicated that these coals would be satisfactory major constituents of coking blends.

Monongalia County, W. Va.

Three coals, one from the Pittsburgh bed and two from the Sewickley bed, ranked as high-volatile A bituminous. Although the physical properties of the coke were satisfactory for coals of their rank, the moderately high sulfur content (2.4 percent) of the Sewickley coals would limit their use as major constituents of coking blends.

Fayette County, W. Va.

Lower Eagle coal, which ranked high in the high-volatile A classification, and Fire Creek coal, which ranked as low-volatile bituminous, had satisfactory chemical properties for metallurgical coals and yielded strong coke. However, coals of these ranks tend to expand excessively when carbonized singly; this factor would limit their metallurgical use.

Raleigh County, W. Va.

A report has been published on the carbonizing properties of Raleigh County coals.[54] Seventeen coals from 13 beds were carbonized singly. To determine the effect of blending the higher rank coals from this county with lower rank coals, five coals were blended with various high-volatile A coals. Raleigh County coals rank as high-volatile A, medium-volatile, or low-volatile bituminous, and this diversity of rank is reflected in the wide variation of the carbonization properties. The higher rank coals yielded the stronger cokes. The blending properties of the low-volatile coals were similar, and the coals can be substituted for each other without significant loss in coke strength of the blends. The expansion of low-volatile coals is not an important factor in blends containing high proportions of high-volatile coal. Although the expanding properties of high-volatile A coals are not closely related to coal rank, the expansion of their blends with low-volatile coal can be estimated from the expansion of the high-volatile component.

Jefferson and Clearfield Counties, Pa.

Three samples of Lower Freeport coal from these counties were high-volatile A and yielded satisfactory coke for coal of that rank. However, the sulfur content of these coals ranged from 2.5 to 3.5 percent; this factor would limit their use as metallurgical coals.

Green County, Pa.

Pittsburgh coal was moderately high in sulfur but could be blended with higher rank low-sulfur coals to yield a satisfactory metallurgical coke.

Indiana County, Pa.

A report has been published on the carbonizing properties of Indiana County coals.[55] Coals in this county rank either high in the high-volatile A or low in the medium-volatile coal classification. Although Indiana County coals produce coke of satsifactory strength for metallurgical use, the variability of their chemical and expanding properties must be considered in their use by the coking industry. The coals contained moderate proportions of ash, but 8 of the 16 coals tested contained over 1.7 percent sulfur, and the maximum was 4.1 percent. The expansion of the coal, as determined in the sole-heated oven, ranged from 19.3-percent contraction to 21.8-percent expansion, therefore, all coals in this county should be tested for expansion before industrial carbonization.

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[54] Wolfson, D. E., Birge, G. W., and Naugle, B. W., Carbonizing Properties of Raleigh County, W. Va., Coals: Bureau of Mines Rept. of Investigations 5359, 1957, 19 pp.

[55] Wolfson, D. E., Birge, G. W., and Naugle, B. W., Carbonizing Properties of Indiana County, Pa., Coals: Bureau of Mines Rept. of Investigations 5314, 1957, 18 pp.

Westmoreland County, Pa.

A report has been published on the carbonizing properties of Westmoreland County coals.[56] One sample from the Redstone, two from the Pittsburgh, and two from the Upper Freeport beds were carbonized in this investigation. Although Westmoreland County coals yield coke with satisfactory physical properties, their use by the coking industry depends on their chemical properties. Four of the coals contained more than 2 percent sulfur. Upper Freeport coals obtained at most locations in the county can be upgraded to meet chemical standards for metallurgical coals, but upgrading of high-sulfur Pittsburgh and Redstone coals would be difficult.

Metallurgical Use of Anthracite

Blends With Bituminous Coal

In recent years economic factors have provided considerable incentive for establishing new markets for anthracite, one possible outlet being in metallurgy, where moderate quantities of anthrafines might be used in coking blends. Studies of the effects of varying the composition of the coal fraction of a blend containing a fixed quantity of anthrafines have been completed.[57] This cooperative work consisted of carbonizing tests in the Tuscaloosa oven of blends in which a number of different high- and low-volatile coals were substituted for those normally used in company coking operations. Comparison of the physical properties of the various cokes indicated several blends that would be expected to yield cokes having physical properties very similar to those produced from the reference blend.

Calcination

Besides being used in blends with bituminous coals, anthracite may be used for blast-furnace and cupola operation, with or without thermal stabilization by calcination.[58] Preliminary foundry tests conducted with anthracite calcined in a vertical-shaft retort resulted in more satisfactory cupola operation than had been obtained with raw anthracite. One-hundred-percent calcined anthracite used as cupola fuel met the foundry requirements for melting rates, temperatures, and fuel-iron ratios. To make further studies of the conditions required to produce maximum quantities of thermally stabilized large-size anthracite and to produce sufficient quantities of fuel for additional foundry tests, a pilot-scale, vertical-shaft calciner having a capacity of 10 to about 500 pounds per hour was constructed (fig. 9).

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[56] Birge, G. W., and Wolfson, D. E., Carbonizing Properties of Westmoreland County, Pa., Coals: Bureau of Mines Rept. of Investigations 5374, 1957, 12 pp.

[57] Gayle, J. B., Eddy, W. H., and Shotts, R. Q., Effects of Substituting Various High- and Low-Volatile Coals in Foundry-Coke Blends Carbonized in the Tuscaloosa and Sole-Heated Ovens: Bureau of Mines, Rept. of Investigations 5307, 1957, 16 pp.

[58] Eckerd, J. W., Buehl, R. C., and Tenney, R. F., The Suitability of Anthracite for Blast-Furnace and Cupola Operation: Pennsylvania State Univ., Min. Ind. Exper. Sta. Bull. 70, June 1957, pp. 17-28.

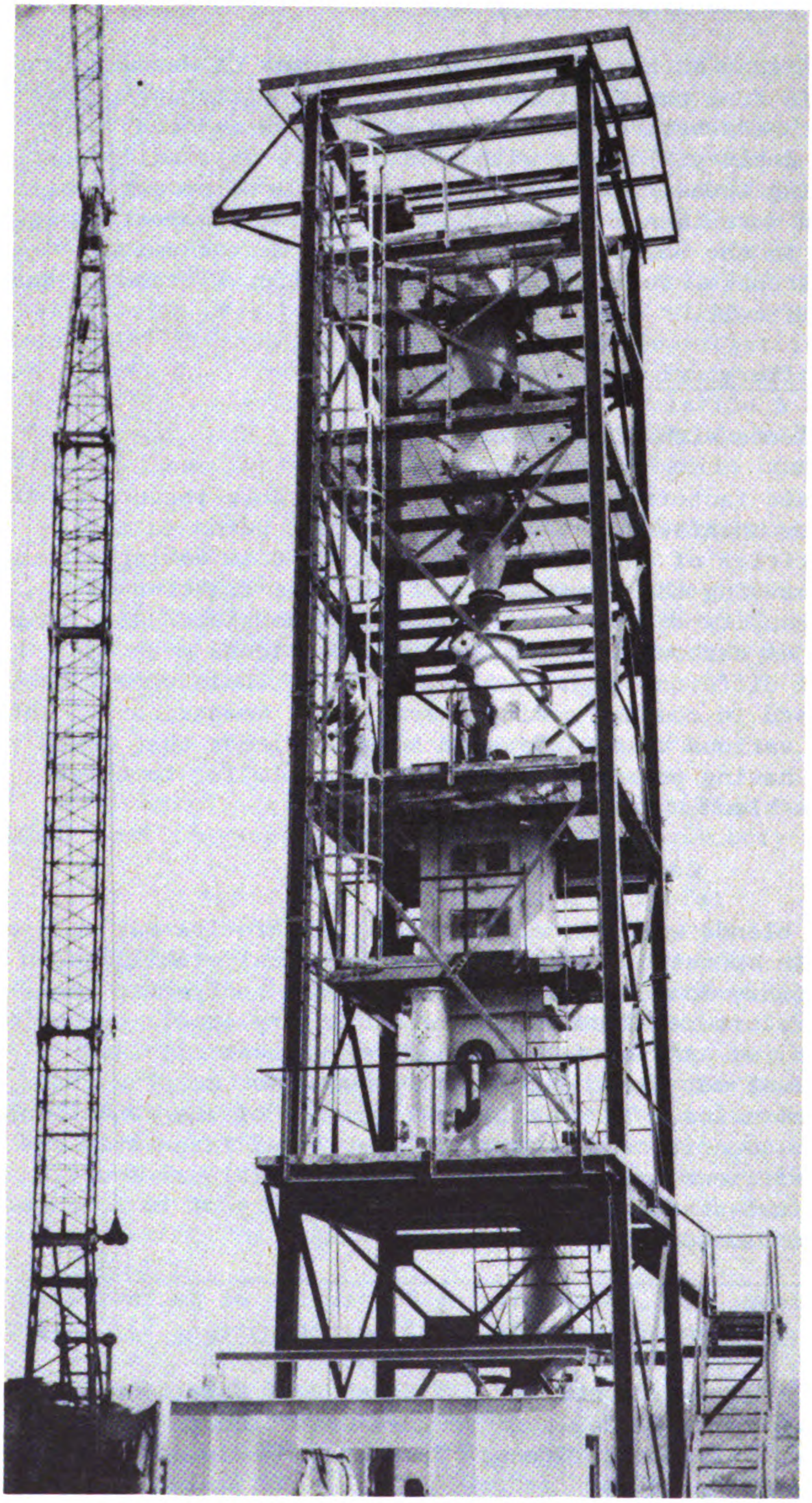

FIGURE 9. - Vertical-Shaft Pilot Plant for Calcining Anthracite.

The calcining characteristics of two types of anthracite have been reasonably well established in this equipment. The first of these, a relatively high-volatile and friable anthracite from the western end of the Western Middle field, has proved to be rather difficult to calcine because it decrepitates to a considerable extent when subjected to either high calcining temperatures (above 1,800° F.) or high throughput rates. When the retort temperature reduced to 1,600° F. and the throughput decreased to about 40 pounds per hour, the yield of plus-1-5/8-inch product increased to 57 percent, but it appeared to be quite friable. In tests with a more stable anthracite from the Northern field throughput rates of approximately 60 pounds per hour were obtained with a yield of 65 to 80 percent of plus-1-5/8-inch product. The product from both anthracites contained approximately 1 percent volatile matter.

To maintain satisfactory operating pressures, the amount of fuel smaller than Stove size (2-7/16-inch by 1-5/8-inch round-hole screens) must be maintained at a minimum in a foundry cupola. In addition, when the ash content of the fuel charge exceeds 12 percent, operating problems are created with regard to both the quality and the amount of slag produced. Satisfactory cupola operation has been achieved only when the fuel contained less than 12 percent ash.

Arrangements have been made to obtain anthracite representing each of the producing areas to study its calcining characteristics. Enough low-ash anthracite will be calcined to make possible further, more extensive foundry tests.

Briquetting

Despite lowering of the thermal decrepitation of anthracite by calcining, its strength as measured by the ASTM Tumbler and Drop Shatter Test for Coke has not been increased materially. To overcome these deficiencies, a laboratory study has been conducted on reconstituting anthracite fines into an agglomerated metallurgical fuel. The work has utilized briquetting, followed by calcination of the preformed product. Coal-tar pitch, asphalt, and sulfite liquor have been evaluated as binders. Two types of bituminous coal were used as mix additives; and effects of size consist, briquetting temperature and pressure, calcination, rate of heating, and final temperatures were evaluated. Briquets formed in a laboratory press at 3,000 p.s.i. and calcined at 1,750° F. were stronger than a good-grade foundry coke. The briquet formulation was 82 percent anthracite, 10 percent bituminous coal, and 8 percent 70° C. m.p. coal-tar pitch. Briquets containing 89 percent anthracite and 11 percent 70° C. m.p. coal-tar pitch were superior in stability factor to metallurgical coke. If this relative strength can be maintained in larger scale production, these blends will be studied as basic metallurgical briquet formulations. A pilot plant is under construction at present to produce large enough quantities of briquets for tests in modern metallurgical equipment and, at the same time, to determine the most satisfactory process conditions. It is expected that the process economics will be at least partly evaluated in the pilot-plant study.

Low-Temperature Carbonization

Wyoming Coals

Production and evaluation of industrial carbons from Wyoming noncoking or slightly caking coals are part of the program concerned with upgrading Western United States coals. Emphasis has been given to the processing of fine and lump coal for the production of char for thermal power, char suitable as a blending material in metallurgical coke manufacture, lump chars that may be substituted for coke breeze in phosphate ore reduction furnaces, and char for manufacturing activated carbons.

Wyoming bituminous and subbituminous coals were processed in bench-scale apparatus to determine potential carbonization yields and in pilot-plant retorts to produce enough char and tar for analysis. These tests have shown that a critical selection of coal is necessary when the coal is processed for a specialized carbon. The type of coal becomes important when carbonization conditions are held constant. Generally, Wyoming bituminous and subbituminous coals produce high yields of tar on carbonization; the yields usually increase as coal rank increases. Characteristically, the low-temperature tars derived from Wyoming coals are high in tar acids. A comprehensive report assessing the carbonizing properties of Wyoming coals is being prepared.

Texas Lignite

A review of recent developments and the status of low-temperature carbonization for generating power was presented before the American Power Conference.59/ The review pointed out that the technical phases of this problem are essentially solved. Economic considerations relating the cost of mining, processing, and yield to the value of solid fuel and tar are complex. These considerations cannot be fully determined until a firm value for low-temperature tar has been established.

Researchers conducted a series of low-temperature carbonization experiments on a dried Texas lignite in the 8-inch-diameter fluid-bed retort, heated both internally and externally, varying the air-lignite ratio. The principal effects of increasing this ratio were a decrease in the yields of char and tar and an increased yield of combustible gas per pound of moisture- and ash-free lignite charged. The decrease of tar is distributed throughout the entire boiling range of the tar, resulting in a significant decrease in tar acids, paraffins, and naphthenes. Gas yields show an increase in all constituents, except hydrogen sulfide, with an increase in transport air. The significant increases, however, may be attributed to carbon monoxide and carbon dioxide. The principal combustible product, accounting for the increase in the gaseous B.t.u. values with increase in transport air, was carbon monoxide. At air-lignite ratios less than 6 standard cubic feet of air per pound of moisture- and ash-free lignite only fixed carbon burned, but at higher ratios portions of both fixed carbon and volatile products burned.

Assay Test

During the past few years interest in low-temperature carbonization of coal has increased, owing to development of new methods, particularly fluidized bed carbonization. The coke or char obtained is a source of smokeless fuel, and the tar is a potential source of raw material for processing into a variety of chemicals. To provide information on the yields of low-temperature carbonization products of American coals, a bulletin was published that gives the results of Fischer-Schrader low-temperature carbonization assays at 500° C. of about 400 coals from nearly every coal-producing State, Alaska, and British Columbia, Canada.60/ The coals ranged in rank from lignite to semianthracite. The report lists the yields of carbonization products, the types of carbonized residue obtained, and the chemical analyses of the coals. The procedure for making the assay is described, as well as a correlation of the yield of carbonization products with the chemical analyses of the coals.

59/ Landers, W. S., Low-Temperature Carbonization of Coal and Lignite for Thermal Power Generation: Proc. American Power Conference, Chicago, Ill., vol. 19, 1957, pp. 344-351.

60/ Selvig, W. A., and Ode, W. H., Low-Temperature Carbonization Assays of North American Coals: Bureau of Mines Bull. 571, 1957, 56 pp.

A different, small-scale, carbonization assay test, originally reported in 1953, is now routinely applied to virtually every coal received by the Denver laboratory. The test has passed through a demonstration period of over 8 years on a wide variety of coals and has adequately proved its versatility, both as a standard bench-scale test and as a research tool in carbonization investigations. Its high reproducibility has shown it to be a most useful guide in predicting plant carbonization yields, particularly from coals of lignitic and subbituminous rank. During 1957 more than 60 new samples were studied.

Utilization of Lignite From India

During 1957 rapid advances were made in the program of technical assistance to the Indian Government in connection with utilization of lignite from Neyveli, South Arcot. The background and scope of this program were reported in 1956. Briefly, the major exploitable reserve of mineral fuel now known in southern India is a deposit of low grade lignite near Madras. Earlier Bureau work, released for publication in 1957,[61] had demonstrated the adaptability of the lignite to fluidized drying and to high-pressure, binderless briquetting. Carbonization of such briquets would provide an alternate source of domestic fuel in India, the demand for which is exceeding the available supplies of dried cow dung and charcoal.

Late in 1956 the Bureau contracted with ICA to pursue further research on the problem and to design and procure an integrated pilot plant for installation at the mine site at Neyveli. These objectives were achieved in 1957. The Neyveli pilot plant has several purposes. First, it processes Indian lignite in large enough quantities to provide solid fuel and tar for use studies on the spot. Second, it serves as a proving ground for later industrial units, the detailed design of which will take advantage of lessons learned in the pilot plant. Third, it will be a facility for training Indian technologists in modern techniques of processing lignite and low-rank coal. The plant has been designed with maximum operational flexibility, so that it can be operated with substantial variations in feed material and in drying, briquetting, and carbonizing conditions.

As a part of the long-range plans for lignite utilization in southern India the Bureau also supervised preparation of an engineering study and preliminary design of a plant (figs. 10 and 11). Capacity of the plant was established as 2-1/2 thousand tons of raw lignite per day, although actual mine production is not scheduled to be this large for several more years. (See also under Foreign Activities, p. 25).

Low-Temperature Tar

Three programs are underway for investigating low-temperature tar. One is a study of lignite tar, another of tars from subbituminous and lower rank bituminous coals, and the third of tar from higher rank bituminous coal.

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[61] Parry, V. F., Landers, W. S., Wagner, E. O., Goodman, J. B., and Allen, R. R., Utilization of South Arcot, India, Lignite: Bureau of Mines Rept. of Investigations 5351, 1957, 34 pp.

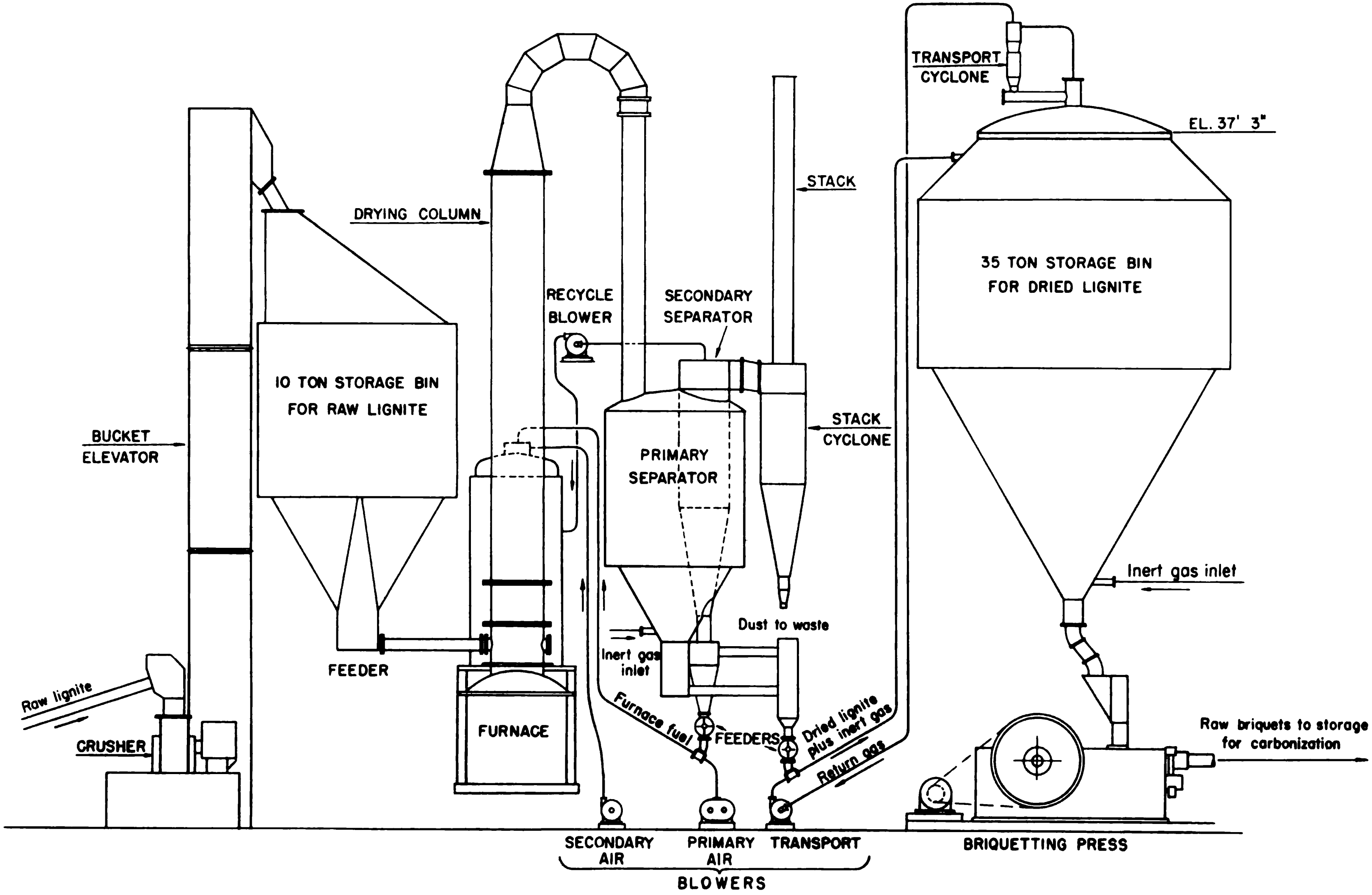

FIGURE 10. - Crushing, Drying, and Briquetting Units for Processing Lignite at Neyveli, India.

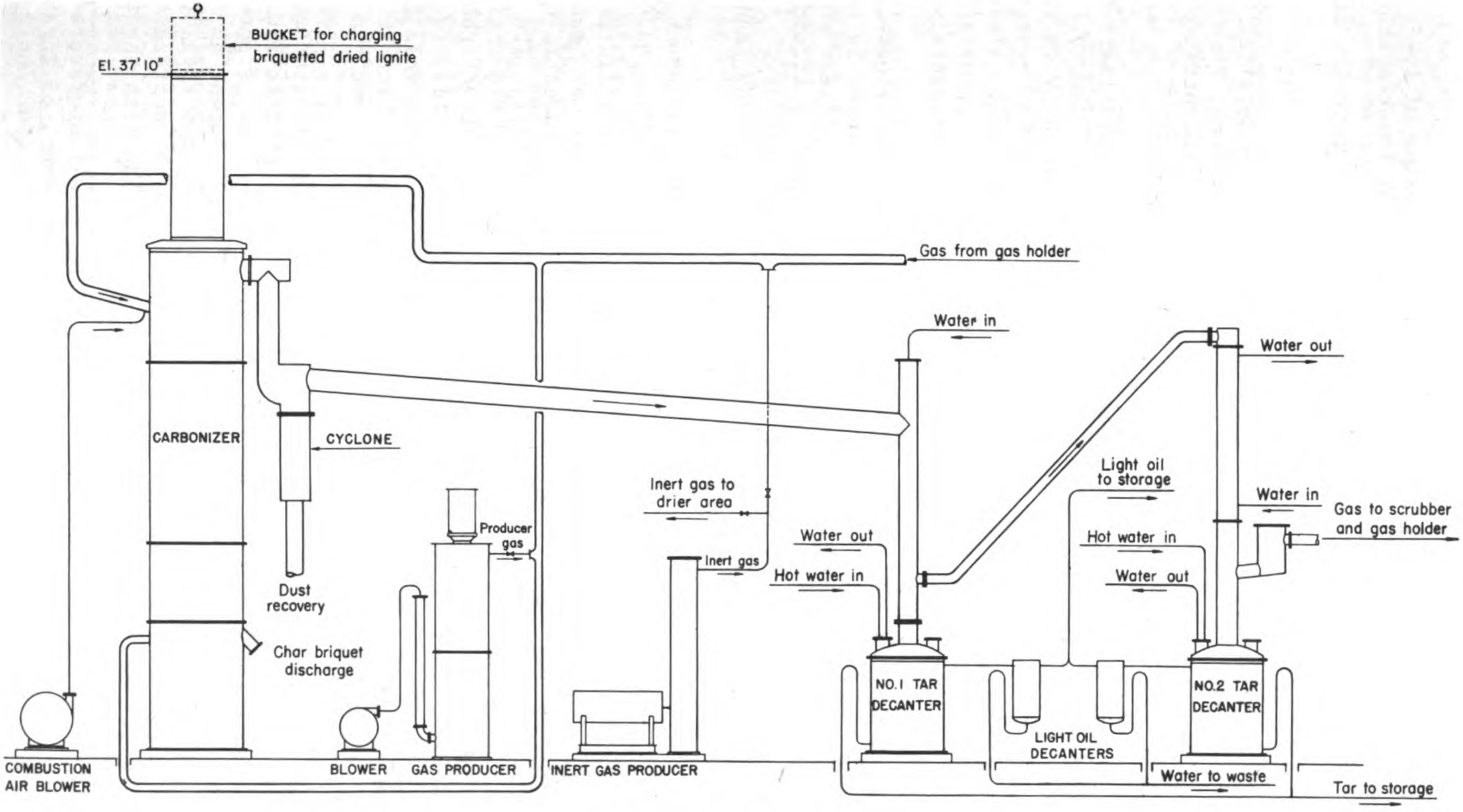

FIGURE 11. - Carbonization and Tar Recovery Units for Processing Lignite at Neyveli, India.

Through this three-pronged attack, it is hoped that the value of low-temperature tar will be increased, aiding economic development of low-temperature carbonization.

Tar From Lignite

The present approach in studying lignite tar is that of detailed characterization of tar fractions by distillation, chemical treatment, chromatography, and examination of absorption spectra. Efforts are being concentrated on the tar acids (phenolics), the more unstable components of the tar.

The most important physical techniques applied to characterization of lignite tar are gas-liquid partition chromatography and infrared absorption spectra. Application of both techniques has been limited essentially to the lower boiling phenolics in the past. The effects of liquid phases and operating conditions on separations of higher phenolics had to be determined. Also, a catalog of reference spectra covering the compounds of particular interest had to be established, necessitating a large amount of laboratory synthesis.

Phenols boiling from 230° to 270° C. have been separated by gas-liquid partition chromatography, but its application to complex mixtures is not yet completely satisfactory; separations of low-boiling phenols present no great problem. The possibility of fractionating tar-acid mixtures by liquid thermal diffusion was investigated, but results have not been satisfactory. A quantitative analysis of the tar acids boiling to 235° C. has been made, and the percentages of 23 individual phenols have been determined.

Tar From Fluid-Bed Carbonizer

One report on the properties of low-temperature tar from a fluidized carbonizer was published during 1957.[62] In later work analyses were made of the tars from fluidized-bed carbonizing tests of 28 foreign and domestic coals varying in rank from lignite to high-volatile B bituminous coal. The 28 coals were from the Western United States, Alaska, Mexico, Ecuador, the Philippine Islands, and India. The samples consisted of six lignites, seven subbituminous C coals, three subbituminous B coals, three subbituminous A coals, two borderline high-volatile C bituminous-subbituminous A coals, two high-volatile C bituminous coals, and five high-volatile B bituminous coals, which show slight agglomerating properties. All coals processed were noncoking and had a heating value of 14,000 B.t.u. per pound (moisture- and ash-free basis) or less. The coals studied had oxygen contents in the range of 11 to 24 percent (moisture- and ash-free basis). Results from the pilot-plant carbonizations indicate that tar acids increase as oxygen increases up to a maximum of 18 percent oxygen; this trend is followed by a decrease in the tar-acid content as the oxygen in the coal increases. If this relationship is confirmed, the total yield of tar acids obtainable from primary low-temperature tar may be correlated with the type of oxygen linkages in the parent coal, rather than with the total oxygen content of the coal. Neutral oil as well as the high-boiling tar fractions increased as total oxygen content of the coal decreased. Both total tar and neutral oil yields increased as heat content in the volatile matter increased.

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[62] Gomez, Manuel, Goodman, J. B., and Parry, V. F., Low-Temperature Tar From Fluidized Carbonizing Reactors: Bureau of Mines Rept. of Investigations 5302, 1957, 44 pp.

Carbonization tests conducted on Texas lignite in the 8-inch-diameter fluidized-bed unit indicate that tar yields increased as the char retention time increased, all other conditions remaining constant. The tar acids and neutral oil fractions of the tar distillate and the olefin fraction of the neutral oil also increased with char retention time. Yields of aromatic and paraffin plus naphthene fractions appeared to be independent of char retention time. In another series of tests a Texas lignite was processed in the fluidized unit at 900° to 1,100° F. Small increases in carbonization temperature increased the specific gravity of the resultant tars and decreased the distillate yield and the temperature of tar decomposition.

Tar From Bituminous Coal

Separation and Characterization

At present not much is known about the many types of chemicals of which low-temperature tars are composed. Therefore, the ultimate value of tar separation and characterization lies in the possibility of supplying industry with information about low-temperature tar in enough detail to make its utilization economically and technologically feasible.

The general program for separating low-temperature coal tar was shown schematically in the 1956 report. This program and the general laboratory and instrumental facilities for composition research have been described.[63/]

During the past year the research on composition of a sample of low-temperature bituminous coal tar progressed far enough so that the first generalities about individual constituents are beginning to emerge. The identities of nearly 100 individual compounds were determined, some on a tentative basis, but most were clearly established. All these compounds fall in the three classes: (1) Tar acids, (2) tar bases, and (3) aromatic hydrocarbons, including benzenes, indanes, indenes, tetralins, naphthalenes, and biphenyls. The tar acids are mostly aromatic in nature, consisting of phenols (including cresols and xylenols), indanols, and naphthols, and the tar bases are also aromatic, consisting of pyridines (including lutidines and collidines), quinolines, and anilines. Because all of these compounds are aromatic, comparisons of molecular structures can be made among the three classes mentioned. In general, the same molecular structures persist through all three classes. The six fundamental aromatic structures are: (1) A benzene-type ring with six carbon atoms; (2) a six-membered aromatic ring with a saturated five-membered ring fused to it; (3) the same structure with one unsaturated, olefinic bond in the five-membered ring; (4) a six-membered aromatic ring with a saturated six-membered ring fused to it; (5) two six-membered aromatic rings fused together to

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[63/] Analytical Chemistry, Laboratory of the Month; Coal Tar Research Challenges Analysts: Vol. 29, No. 1, January 1957, pp. 51A-53A.

form a naphthalene-type structure; and (6) two six-membered aromatic rings joined together by a single bond to form a biphenyl-type structure. This information indicates that the same complexity of structure apparently persists throughout all classes of compounds in the tar, and that a particular structure found in one class of compounds will likely exist in other classes.

Because of the obvious economic significance of phenols and the relative ease with which they can be separated and analyzed, initial work centered mainly around this class of tar constituents. A list of 183 low-boiling phenols and their physical properties was published.[64]

To study the sorption of individual pyridine bases by cation-exchange resins, an accurate and relatively quick method was needed for determining small quantities of these bases in aqueous methanol solutions.[65] This ultraviolet spectrophotometric method can be applied to dilute solutions of individual tar bases and simple mixtures of tar bases.

Upgrading and Utilization

Low-temperature coal tar is an extremely complex mixture of organic compounds; as such, it has little more than fuel value. To increase its value, the whole tar must be separated into its components, some of which must be converted to substances of greater commercial importance. The upgrading plan comprises a series of selected physical separation processes, such as distillation and solvent extraction, and chemical conversion processes, such as dealkylation and oxidation.

A process for isolating tar acids, employing monoethanolamine as the extractant was reported in 1956. Further laboratory studies of this process resulted in development of an alternative procedure for recovering the tar acids from the monoethanolamine solution. The original procedure used carbon dioxide gas to spring the phenols from the amine solution. The amine carbonate formed in this operation was regenerated by thermal decomposition and reused. The alternate procedure consisted of diluting the amine solution with water and extracting the tar acids with a volatile solvent, such as benzene or butylacetate. This alternate procedure gave a product free of monoethanolamine and eliminated the step of thermal decomposition of the amine carbonate.

Studies on separating neutral tar-oil fractions into chemical types by means of solvent extraction were made. Three selective solvents, furfural, diethylene glycol, and dipropionitrile, were investigated. Of the three, dipropionitrile is a relatively new solvent and has high selectivity for aromatic hydrocarbons. Neutral oils boiling at 200° to 250° C. were separated into aromatic and nonaromatic fractions. The aromatic fraction may be used as a feed material for producing phthalic anhydride, which is of great commercial value at present. The nonaromatic fraction, being rich in cyclic compounds, may be used as a blending material for jet fuels. For the same amount of solvent used in single-stage extraction furfural extracted more aromatics than the other two, but the aromatics contained large amounts of nonaromatic hydrocarbons. The reverse was found true for dipropionitrile; that is a product of high quality but smaller in quantity was obtained. Diethylene glycol lay between the two in performance.

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[64] Karr, Clarence, Jr., Physical Properties of Low-Boiling Phenols, a Literature Survey: Bureau of Mines Inf. Circ. 7802, 1957, 15 pp.

[65] Chang, Ta-Chuang Lo, and Karr, Clarence, Jr., Spectrophotometric Determination of Small Quantities of Some Individual Pyridine Bases by Successive Extractions: Anal. Chem., vol. 29, No. 11, November 1957, pp. 1617-1619.

An analytical scheme was devised (fig. 12) to separate the neutral oils into five types: Aromatics, straight-chain olefins, straight-chain paraffins, branched-chain and cyclic olefins, and branched-chain and cyclic paraffins. By repeatedly washing with dipropionitrile nearly all the aromatics and the coloring substances can be removed.

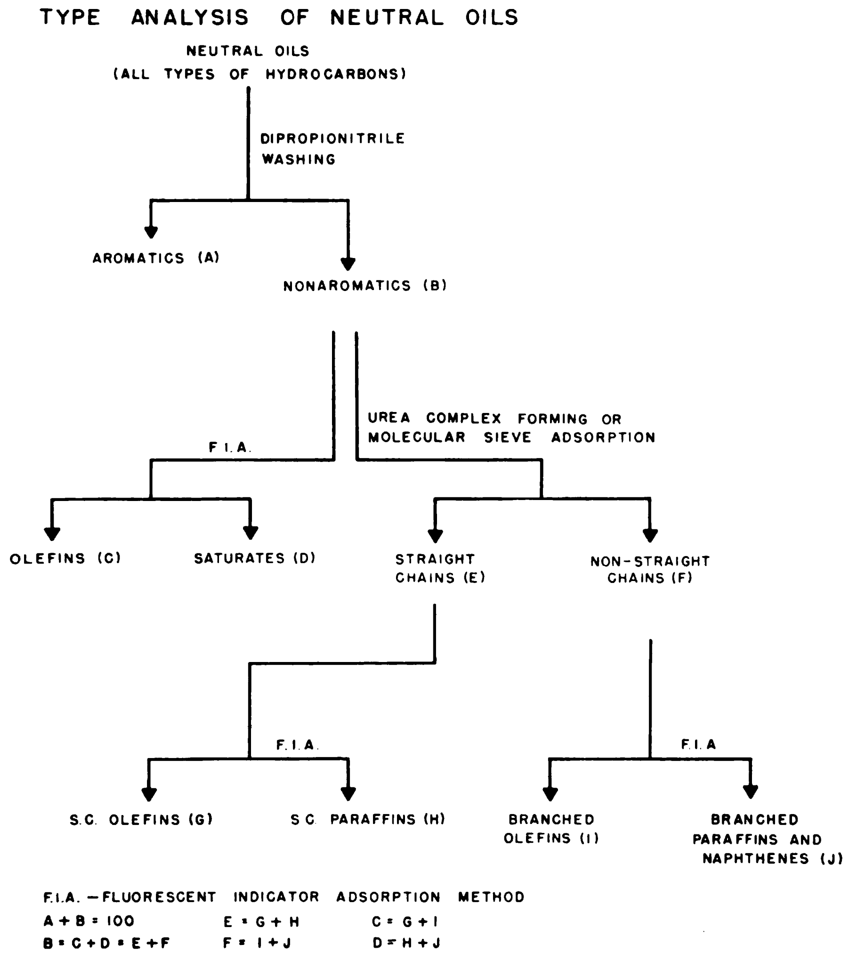

FIGURE 12. - Scheme for Separating Neutral Oils Obtained From Low-Temperature Tar.

As a guide in processing tar acids and tar bases, a comparative study was made of the composition of such mixtures isolated from four low-temperature tars. In these low-temperature tars, tar acids boiling above the range of xylenols represented 75 percent of the total compared with 25 percent for coke-oven tars. The sum of pyridines and quinolines found in these tars was less than 0.5 percent of the whole tar, and the pyridine content was four times as great as the quinoline content.

GASIFICATION OF COAL

Reaction of Carbon With Steam

In gasification of coal the rate is probably limited by the rate of reaction of steam with carbon. Study of the kinetics of this reaction should therefore contribute basic information applicable to the development and design

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of commercial-scale gasifiers. Two experimental methods have been used to investigate this subject: The falling-particle method[66] and a method in which a carbon sample is suspended from an analytic balance in a furnace maintained at a constant temperature ranging from 1,800 to 2,400° F., the rate of weight loss being a measure of the rate of reaction. Over 200 runs have been made by the second method to study the effect of the velocity of steam through the reactor and the effect of partial pressure of steam in the steam-helium atmosphere at various temperatures.

Because reports in the literature have indicated that the steam-carbon reaction is catalyzed by a strong neutron flux at room temperature or by gamma radiation, hollow graphite cylinders containing a 1-curie source of encapsulated cobalt-60 were suspended from the balance, and the rate of reaction with steam was compared with that of similar cylinders containing no cobalt. A series of runs at 1,000° and 1,500° F. showed that gamma radiation had no effect on the rate of reaction under these conditions.

Hydrogen by Steam-Iron Process

The production of hydrogen by the reaction of steam with iron has been employed commercially on a small scale for many years. The iron oxide produced in the oxidation step is reduced with producer gas for subsequent use in the oxidation step. These reactions are carried out alternately with a fixed bed of iron granules or spheres. As only a surface layer of the iron takes part in the reaction, only a few percent of the iron is utilized. Thus, large reaction vessels are required, and investment costs are high. Operating costs also are high because conversion of the reacting gases is low. To increase the extent of reaction and thus improve the economics of the steam-iron process, use of a fluidized bed of solids is being investigated for the reaction. Iron oxide particles of 20- to 100-mesh size are employed. The raw materials are producer gas and steam. Iron ore, preferably magnetite, is used to replace losses of iron oxide. Producer gas is used for reducing the iron oxide because it is the cheapest gas that can be made from coal.

A 4-foot bed of synthetic ammonia catalyst of 20- to 100-mesh size was used, and the linear velocity of the gas was approximately 0.6 foot per second to maintain fluidization. For a given ratio of solids-to-gas flow the conversion of the reducing gases was increased by increasing pressure. As the solids flow was increased at a constant flow of reducing gas, the conversion increased, but the extent of reduction of the iron oxide decreased. For the 4-foot bed of catalyst employed conversion of the reducing gases ranged from 40 to 70 percent, depending upon the solids-gas ratio. To achieve conversion of the gases and a considerable amount of reduction of the solids, a higher bed of solids is necessary than was used in the pilot plant. The rate of conversion of steam as a function of solids-gas flow during oxidation was similar to that during reduction. However, the rate of oxidation is more rapid than that of reduction. Investigation of rates of reaction in the process is being continued to find the optimum conditions of operation.

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[66] Dotson, J. A., Holden, J. H., Koehler, W. A., Rate of the Steam-Carbon Reaction by a Falling-Particle Method: Ind. Eng. Chem., vol. 49, No. 1, January 1957, pp. 148-154.

Gasification of Lignite

Because of its inherently high reactivity, lignite appears to be an especially suitable raw material for gasification.

Results were published on experiments carried out in a small-scale, externally-internally heated annular retort gasifier.[67] The gasifier, having a nominal capacity of 40 pounds of lignite per hour, was operated to provide information on the effect of temperature distribution and supplementary internal heating with air on gas-production capacity. Results were compared with those from a commercial-scale gasifier of similar design. A crude ammonia synthesis gas with a $(H_2+CO):N_2$ ratio of 2.9 was produced at hourly rates up to 164 cu. ft. per sq. ft. of externally heated surface.

In developing a process for gasifying lump fuel in a fixed-bed gasifier with oxygen and steam at elevated pressure, a pilot plant was designed and contracts for equipment were made (fig. 13). The structural steel framework has been erected, and numerous items were received for the final assembly. Calculations on the thermodynamics of pressure gasification have been completed for both lignite (low-temperature) char and as-received lignite. Conditions for which calculations were made included pressures of 10 to 30 atmospheres and assumed equilibrium temperatures of 1,000° (1,340°) to 1,300° K. (1,800° F.). Oxygen-steam ratios ranged from 0.1 to 1.9 mol. per mol. (2.1 to 39.9 cu. ft. per lb.), depending upon the overall thermal requirements for the reactions considered. High equilibrium temperatures give high (H_2+CO) concentrations in the product gas and relatively low exit-gas temperatures. Provided equilibrium can be approached at the higher temperatures, oxygen-steam ratios of 0.7 to 1.0 mol. per mol. appear favorable for use with char. Somewhat higher ratios can be used with as-received lignite due to the thermal requirements of drying and carbonizing the fuel.

Bench-scale carbonization tests have been completed for evaluating the agglomeration and carbonization characteristics of as-received and steam-dried lignite and subbituminous coal at a controlled temperature of 945° F. and at pressures ranging from atmospheric to 1,000 p.s.i.g. of hydrogen. To evaluate the effect of hydrogen on the carbonization characteristics, experiments were also run in a nitrogen atmosphere under similar test conditions. Agglomeration of feed material was not found to be a problem under conditions expected in the gasifier. Some coke formation was noted with lignite in a hydrogen-rich atmosphere at pressures from 600 to 1,000 p.s.i.g., and maximum agglomeration was at 1,000 p.s.i.g. However, all agglomerates had low mechanical strength and could be crushed easily between the fingers. Lignite agglomerated more than the subbituminous coals that suffered less thermal decomposition and partial hydrogenation. Yields of gas and solid residue followed the general pattern, because more gas was formed at the higher pressures, whereas yield of solid residue decreased. Tar yield was highest at 1,000 p.s.i.g.; at the lower pressures, change was insignificant.

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[67] Oppelt, W. H., Kamps, T. W., Gronhovd, C. H., Kube, W. R., and McMurtrie, R., Production of Crude Ammonia-Synthesis Gas From North Dakota Lignite in an Annular-Retort Gasifier: Bureau of Mines Rept. of Investigations 5297, 1957, 29 pp.

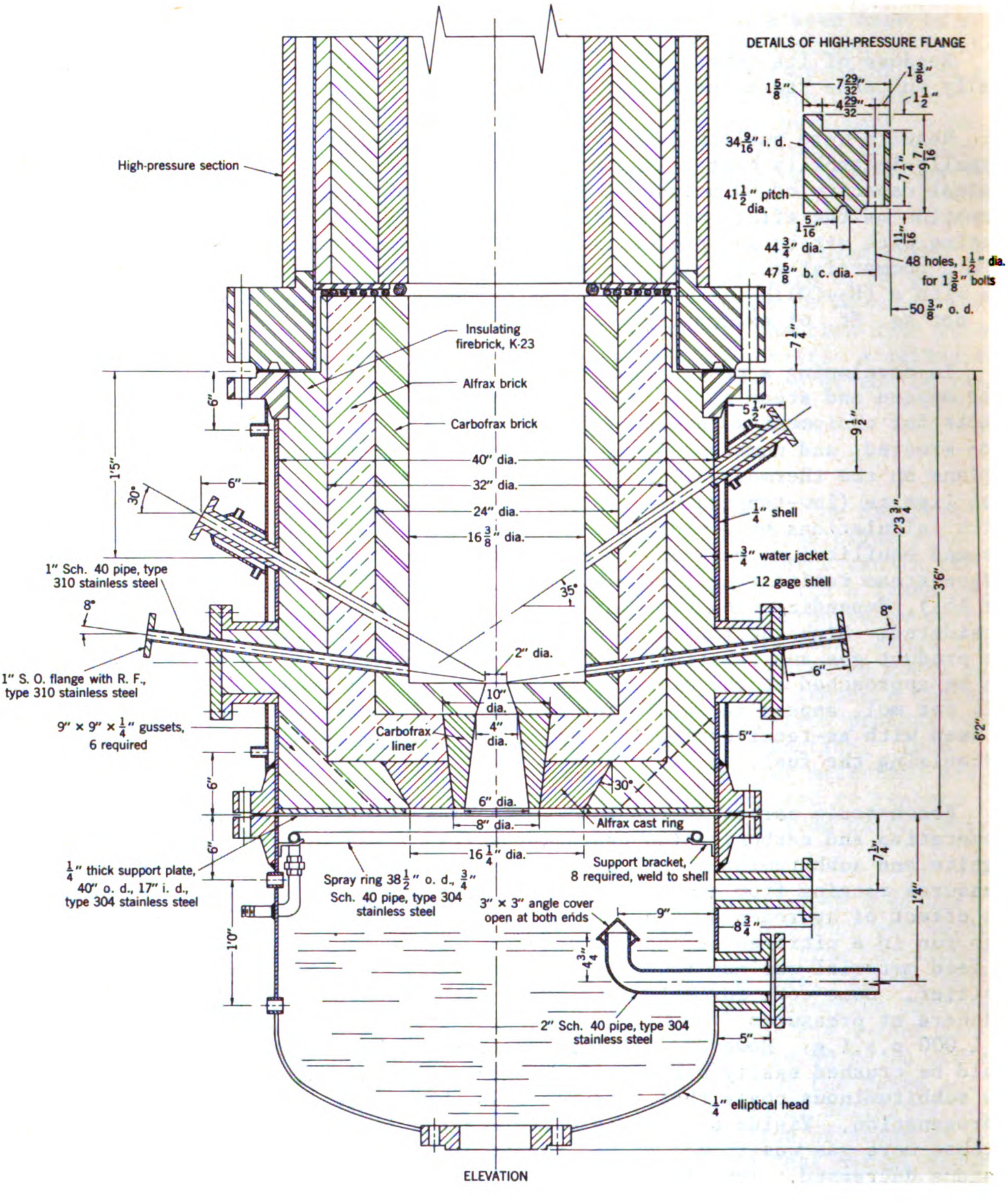
DETAILS OF HIGH-PRESSURE FLANGE
1 5/8"
7 29/32"
4 29/32"
1 3/8"
1 1/2"
34 9/16" i. d.
7 1/4"
9 7/16"
41 1/2" pitch dia.
1 5/16"
11/16"
44 3/4" dia.
47 5/8" b. c. dia.
48 holes, 1 1/2" dia. for 1 3/8" bolts
50 3/8" o. d.
High-pressure section
Insulating firebrick, K-23
Alfrax brick
Carbofrax brick
7 1/4"
6"
5 1/2"
9 1/2"
1'5"
30°
6"
40" dia.
32" dia.
24" dia.
16 3/8" dia.
1/4" shell
2'3 3/4"
3/4" water jacket
12 gage shell
3'6"
6'2"
1" Sch. 40 pipe, type 310 stainless steel
35°
8°
8°
2" dia.
1" S. O. flange with R. F., type 310 stainless steel
6"
10" dia.
4" dia.
9" × 9" × 1/4" gussets, 6 required
Carbofrax liner
5"
6"
30°
6" dia.
8" dia.
Alfrax cast ring
6"
16 1/4" dia.
7 1/4"
1/4" thick support plate, 40" o. d., 17" i. d., type 304 stainless steel
Spray ring 38 1/2" o. d., 3/4" Sch. 40 pipe, type 304 stainless steel
Support bracket, 8 required, weld to shell
1'4"
3" × 3" angle cover open at both ends
9"
8 3/4"
1'0"
4 3/4"
2" Sch. 40 pipe, type 304 stainless steel
5"
1/4" elliptical head
ELEVATION

FIGURE 13. - Low-Pressure Slagging Section of Fixed-Bed Gasifier.

Gasification of Pulverized Coal

Pilot-Plant Operation

For several years gasification of pulverized coal to produce gas for synthesis of hydrocarbons, methanol, or ammonia has been studied on a pilot-plant scale.[68] Changes in design have been introduced periodically to improve operations. Recently, a new refractory lining was installed in the reaction zone of the pressure gasifier. It changes the heat conductivity through the lining and permits evaluation of the effect of this variable on gasification.

Coal-Feeding Methods

Work in developing methods of feeding pulverized coal into a pressure gasifier continued with emphasis on a system in which a slurry of 50 percent pulverized coal and 50 percent water is heated to yield a mixture of superheated steam and preheated coal. Preheating lowers the requirement of oxygen, the third medium of gasification. However, before such a mixture can be fed into a pressure gasifier, its steam-coal ratio must be reduced. A suitable steam separator is being developed.

Some work has also been done on a system for feeding steam and coal in which pulverized coal under pressure is inspirated into a stream of steam, which then passes through a heater into the pressure gasifier.

Modification of the fluidized feeder used in the pilot plant from a "single-zone" to a "two-zone" feeder" feeder (fig. 14) allowed lowering the fluidizing gas requirements to 25 percent of the requirements for a single zone. Carryover of particles (coal entrainment) was reduced to a trace and the feeding capacity of the fluidizer doubled.

In this "two-zone" feeder the distribution plate, normally used for fluidization in the large feeder, was altered so that a central column and an annular column could be fluidized at different rates. A central column with a large funnel at the top was placed above the altered plate. In operation, the inner column of coal is fully fluidized, and the coal feeds out through the small funnel and extraction pipe. The annular column is only slightly fluidized. The large funnel forces gas from the annular column to the outer edge of the feeder and assists in feeding the coal from the upper zone down into the small fluidized column.

Externally Heated Reactor

Because gasification of coal by steam is an endothermic reaction, heat must be added to the system to maintain the rate of reaction. In a pressure gasifier the necessary heat is generated internally by adding oxygen that reacts with the coal to the steam required for gasification. Heating the reactor

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[68] Strimbeck, G. R., Holden, J. H., McGee, J. P., and Hirst, L. L., Recent Work by the Bureau of Mines on the Gasification of Pulverized Coal: American Gas Association, Operating Section (Preprint) CEP-55-27, 32 pp.

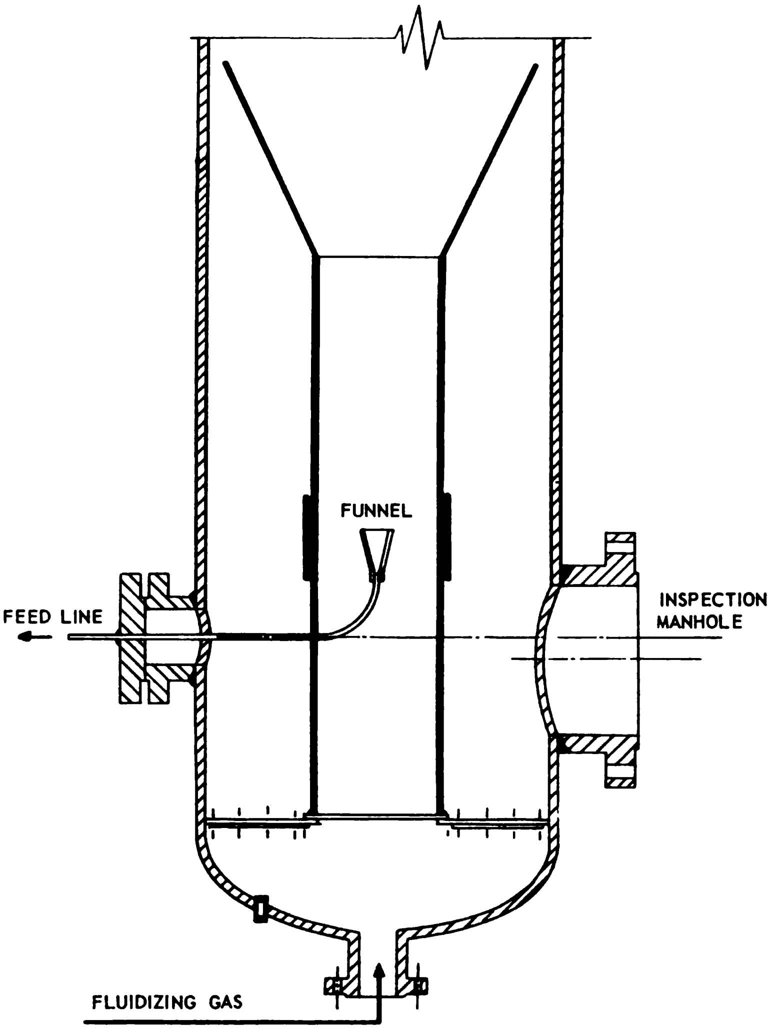

FIGURE 14. - Section of Two-Zone Feeder for Feeding Powdered Coal to a Gasifier.

externally is an alternative method, and about 80 runs in an externally heated coil have demonstrated that gasification of coal-water slurries fed to such a reactor is feasible (fig. 15). The coil was kept at 1,500° F. or above continuously for 1 year, and coal-water slurries were fed through it intermittently for a total time of about 100 hours. The coil was in good condition at the end of this period. The younger, more reactive coals gasified more completely and at a faster rate. Over 90-percent carbon conversion has been obtained with a geologically older (higher rank) lignite from Lake DeSmet, Wyo., at 1,900° F. in less than 1 second residence time. Longer residence times and higher reaction temperatures increased the carbon conversion to gas but had little effect on the gas composition. The average gas composition was 57 percent H_2, 20 CO_2, 15 CO, 4 CH_4, 2 unsaturated hydrocarbons, and 2 percent N_2. A linear relationship was found between carbon conversion and the ratio of steam to coal: The greater the amount of steam present, the more carbon was gasified under similar conditions.

Nuclear Energy for Coal Gasification

Energy from a nuclear reactor may be used in three ways that are of interest in coal gasification: (1) The energy released as heat may be used directly in a gasification unit to maintain a high rate of gasification; (2) the heat may be used indirectly through some heat-transfer medium to accomplish the same objective; and (3) the energy may be used to superheat steam to such a high temperature that the reactions are self-sustaining.

In 1955 a cooperative program with AEC was initiated to develop nuclear reactor technology to permit heating of gases to 2,500 to 3,500° F. and to investigate methods of applying this heat to a steam-coal mixture so that

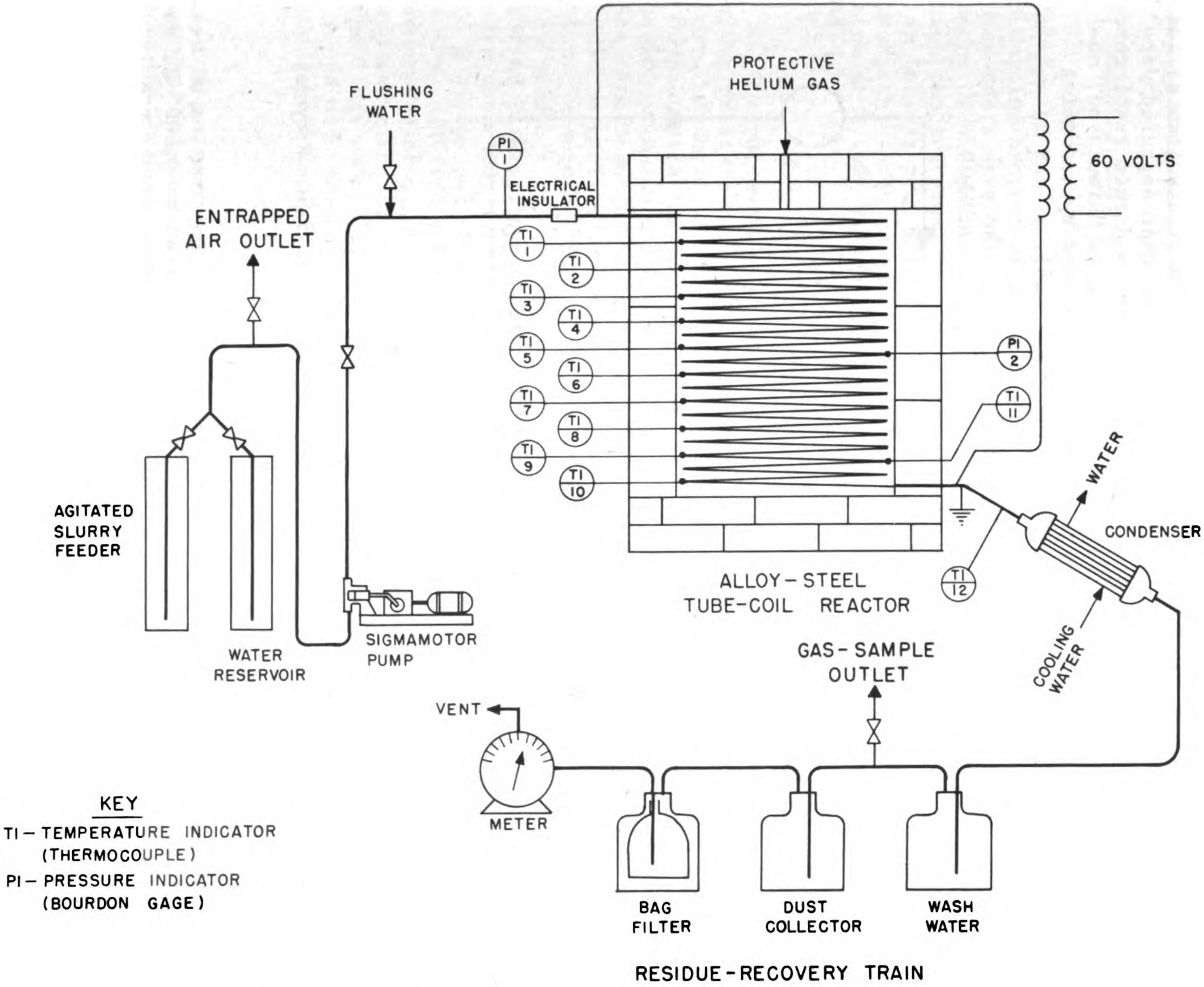

FIGURE 15. - Externally Heated Coil for Gasifying Coal-Water Slurries.

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essentially no radioactivity is imparted to the products. Because irradiation tests on coal and coal ash showed that the ash became radioactively "hot," even after 30 seconds exposure, the indirect method was selected with helium as the heat-transfer medium. An alternative gasification process that appeared very attractive was to superheat the steam in a nuclear reactor and then react the steam with coal in a separate unit. To study this problem, a "simulated" nuclear reactor has been built in which graphite spheres heated by induction serve as counterparts for the nuclear fuel elements (fig. 16).

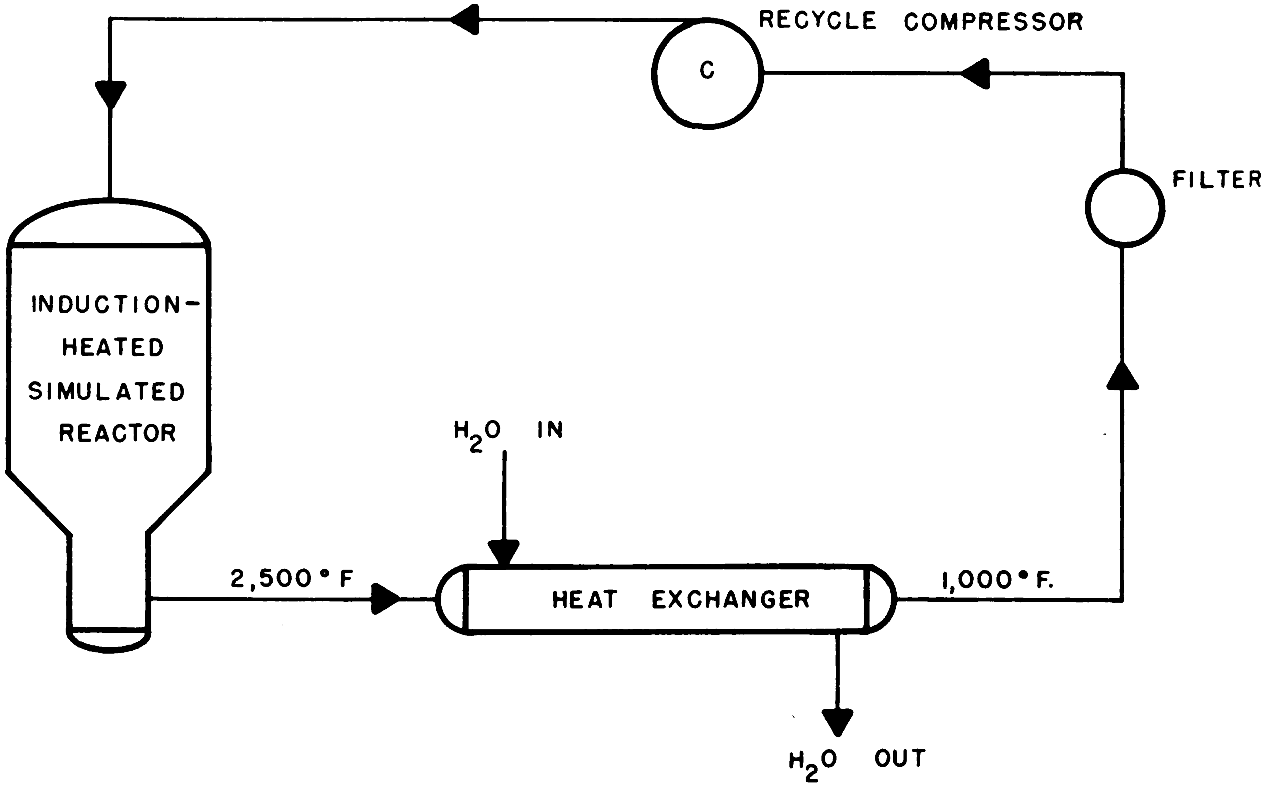

FIGURE 16. - Simplified Flowsheet for Simulated Nuclear Reactor With Helium Recycle.

Heat Transfer

In a first step, this reactor has been used to procure information on heat transfer by helium at atmospheric pressure. The gas is heated to 2,500° F. and cooled in a gas-to-gas heat exchanger to about 1,000° F. Experiments are planned at pressures up to 250 p.s.i.g. Both the compressor and the heat exchanger require development programs. Preliminary designs have been made and a prototype compressor and heat exchanger have been ordered. Some heat-transfer measurements have already been made at low rates of flow of helium because of the low capacity of the compressor used to circulate the helium. Two types of ceramic heat exchangers have been designed for the temperature range 2,600° to 2,800° F. and were fabricated of vitreous alumina. One is a shell-and-tube exchanger fabricated in one piece. The other design permits the installation of thermocouples at 3-inch intervals along the interior tube surface followed by the final cementing of joints in place. This heat exchanger has been in satisfactory operation for 2 months at a variety of flow conditions and at temperatures as high as 2,500° F.

Materials for Construction

Laboratory tests are in progress on materials required for effective utilization of nuclear heat. Material-testing reactors have been constructed to determine properties and reactions of materials at high temperatures. Endurance tests have been made to determine the heat, corrosion, and erosion resistances of various ceramics and cermets. Numerous materials have been tested at high temperatures, but no ideal cladding material has yet been found for coating ball-shaped, graphite fuel elements containing fissionable materials. However, the technical feasibility of using unclad graphite fuel elements in helium at 2,500° F. has been demonstrated under simulated nuclear conditions.

Gasification of Anthracite

For successful operation of the Lurgi gasification process, which operates under pressure with steam and oxygen, only noncaking coals have been satisfactory. Anthracite fulfills these requirements; hence, two tests - one on Chestnut size anthracite containing 27 percent ash and the other on a Rice-Buckwheat mixture containing 19 percent ash - were made at the Dorsten works of Steinkohlengas A. G. in Germany. The tests demonstrated that both samples could be gasified. Although optimum operating conditions could not be determined because of limited oxygen supply and because it was necessary to maintain the producer in satisfactory operating condition at all times to provide gas for an international gas grid, reliable data within these operating limitations were obtained. The highest throughput rate obtained was 273 pounds of coal (dry basis) per square foot per hour or 199 pounds per square foot per hour on a dry, ash-free basis. In general, as the throughput rates increased, the carbon-conversion rate decreased; hence, the combustible residue in the ashes increased with the higher rate, owing, primarily to the limited oxygen supply and the large amount of steam used for the operation. Bureau of Mines observers agreed with the Lurgi engineers that certain changes would have to be made to the gasifier to take advantage of the high ash-fusion temperature of anthracite.

Underground Gasification

Experiments in underground gasification of coal are in progress to develop methods whereby chemical products or thermal energy from unmined coal may be brought to the surface. Such a method might permit utilization of coal in beds that cannot be profitably mined or of coal that remains in areas where mining operations have been completed.

The tests completed have been conducted in Alabama in the America coalbed, which is double bedded, heavily cleated and about 5 feet thick, including 10 to 11 inches of fire-clay parting. A permeable path is developed by hydraulic fracturing and injection of fluids in the coalbed. This path is connected to air and gas piping on the surface by boreholes. Gasification paths are then developed between the boreholes by a procedure called "backward burning," in which the coal is ignited some distance from the air-injection hole, and partial combustion then proceeds in a comparatively straight line toward the point of air injection until the path is linked by a partly devolatilized and relatively permeable mass of coal. Then the air flow is reversed and gasification of the coal occurs.

In the most recent hydraulic fracturing experiment the primary objective was to propogate and extend a fracture in the coalbed by means of water loaded with larger sand grains than had been used in the first experiment, in which kerosine and finer sand were tried. During postfracture permeability testing, it was discovered that the inflow and outflow capacities of the boreholes (except the fracture injection borehole) could be increased significantly by injecting 100 to 1,000 gallons of water. This suggested that, properly applied, small amounts of water might eliminate the expense of using equipment for injecting large amounts of fluid and sand.

Gasification operations and results in this second fracturing experiment were similar to those previously obtained. The quality of gas produced initially reached a maximum of 170 B.t.u. per cu. ft. In the following 2-week period the gas quality varied between 100 and 120 B.t.u. per cu. ft. During the next 2-week period the gas quality gradually declined to 50 B.t.u. per cu. ft.

A series of small-scale electrolinking tests in the Pratt coalbed at a nearby site were completed in September 1956 and included seven trials in which vertical electrodes were used similar to those used in the large-scale work.[69] Six of these linkages were exposed by mining operations during July 1957. Figure 17 shows the type of linkage obtained - generally cylindrical, about 7 inches in diameter, and not straight. It contained slag or dense coke in the center that changed gradually to a soft coke at the perimeter. Numerous radial and horizontal cracks or fissures in the linkage allowed air or gas to pass from one end to the other. The measured length of the linkage and the time of carbonization were related to the time required for linking and the power consumed during carbonization.

Gas Purification

Commercial utilization of gas made from solid fuels is possible only when the components are in a suitable ratio and when the gas is relatively free of such impurities as carbon dioxide, sulfur compounds, and solids. Naphthalene may also be troublesome.[70]

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[69] Elder, James, L., Fies, M. H., Graham, Hugh G., Capp, John P., and Sarapuu, Erich, Field-Scale Experiments in Underground Gasification of Coal at Gorgas, Ala. Use of Electrolinking-Carbonization as a Means of Site Preparation: Bureau of Mines Rept. of Investigations 5367, 1957, 101 pp.

[70] Kane, L. J., Gentile, J. J., Trostle, K. E., Katell, S., and Shale, C. C., Turbidimetric Determination of Naphthalene in Gas: Bureau of Mines Rept. of Investigations 5368, 1957, 12 pp.

Removal of Gases

When coal is gasified with oxygen and steam to produce a mixture of carbon monoxide and hydrogen, a considerable quantity of carbon dioxide is formed, along with smaller amounts of hydrogen sulfide and carbonyl sulfide from the sulfur in the coal. Because Fischer-Tropsch catalysts are poisoned by an extremely small amount of sulfur and their activity is impaired by high concentrations of carbon dioxide, these impurities must be removed before synthesis.

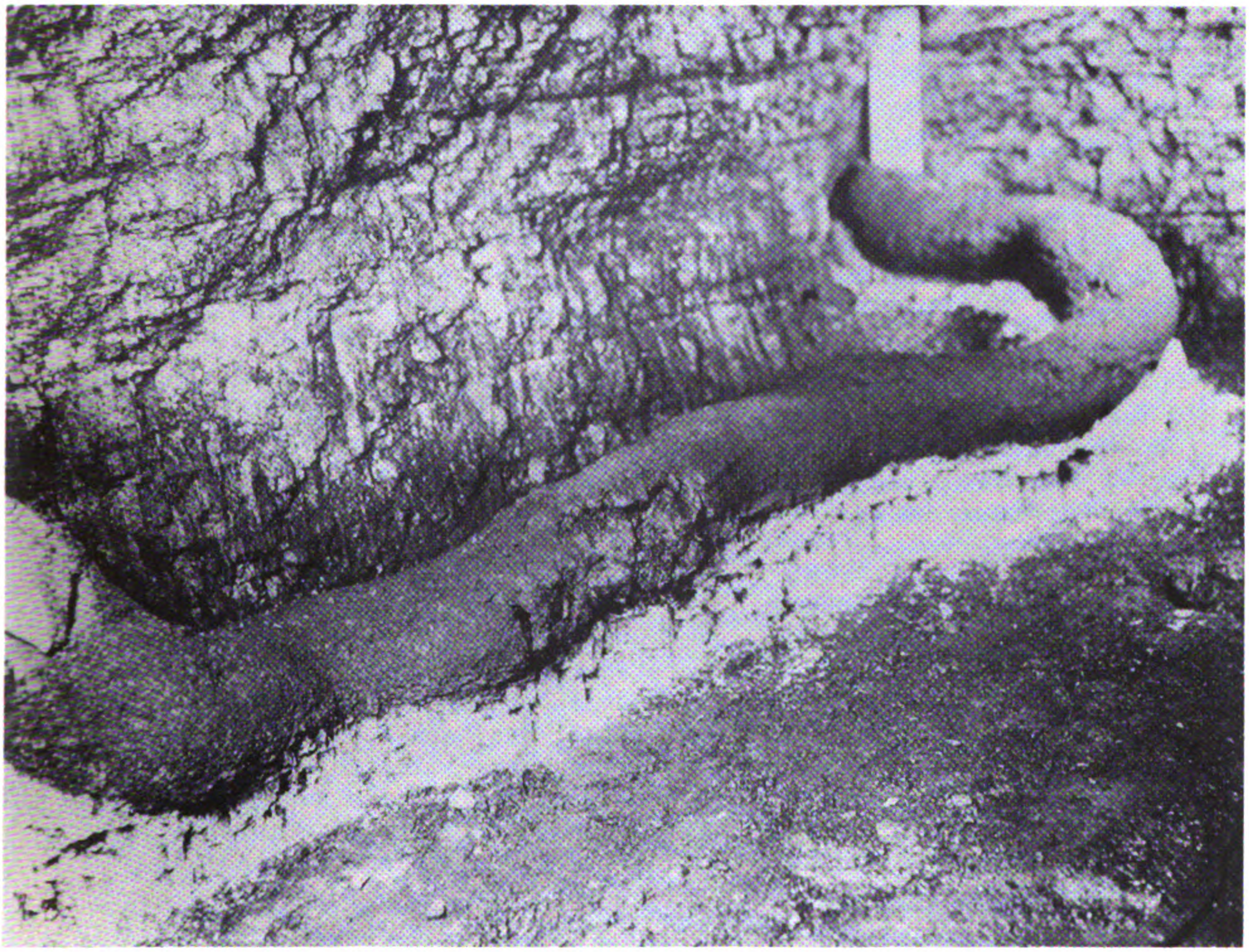

FIGURE 17. - Section of Coal Carbonized Underground During Electrolinking.

Purification of synthesis gas by conventional processes is expensive. Purification by a hot solution of potassium carbonate at elevated pressure has been under study in a pilot-plant unit. Because absorption takes place at an elevated temperature, no heat-exchange equipment is necessary between the absorption and regeneration steps, as in the conventional processes.

Bench-scale investigation was continued to determine equilibrium pressures of hydrogen sulfide and carbon dioxide over hot solutions of potassium carbonate containing various concentrations of these two gases. During the past year emphasis has been placed on pilot-plant operations to determine the extent to which hydrogen sulfide can be removed without affecting the steam consumption for regeneration. Selection of optimum operating conditions may permit elimination of iron oxide boxes to remove the final traces of hydrogen sulfide.

Because carbonate solutions are corrosive, a program is underway to determine the extent of their attack on various metals and the degree to which common inhibitors will eliminate such chemical action. Attack by uninhibited carbonate solution, acidified with carbon dioxide to a carbonate-bicarbonate ratio of 0.34, resulted in severe metal loss on both high- and low-carbon steel. Brass and stainless steel were relatively unaffected. Results of tests with unacidified solutions were similar for each metal tested, except that the degree of attack was much less severe.

Removal of Dust

Dust must be removed from the gas stream entering a fixed catalyst bed to avoid loss of catalytic activity. Tests have shown that accumulation of dust in the bed decreases the available surface area of the catalyst. Periodically a mass of dust may break loose and pass through the bed, restoring the activity, in part. Little or no dust penetration into the surface of catalyst pellets has been found.

The effect of dust depends both on its concentration and on its size distribution. Methods are being developed for generating hot dust-laden gas, for sampling that will not cause agglomeration of dust particles and thus false size distributions, and for removing dust at elevated temperatures and pressures. For sampling a fine wire-mesh filter has been found satisfactory. Studies are underway to develop a high-temperature electrostatic precipitator. Theoretical considerations indicate that a magnetic field superimposed upon the electrostatic field in the precipitator should improve the removal of dust at elevated temperatures, and this possibility is being investigated.

Process Economics

As a guide for its research programs, both completed and underway, the Bureau makes cost studies of the processes and unit operations involved. Two were published this year.[71 72]

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[71] Katell, Sidney, and McGee, J. P., An Economic Study of Air Compression Costs: Cost Engineering, vol. 2, January 1957, pp. 5-9.

[72] Katell, Sidney, and Faber, John, An Economic Evaluation of Cooling Water Costs: Cost Engineering, vol. 2, No. 3, July 1957, pp. 70-76.

COMPLETE CONVERSION OF COAL TO FLUID FUELS

Coal Hydrogenation

Autoclave Studies

The mechanism of the iron-catalyzed hydrogenation of coal was studied in a rotating batch autoclave at 450° C. and hydrogen at 8,000 p.s.i. (fig. 18). Experiments were conducted with a paraffinic vehicle (n-octadecane), with coal-hydrogenation oil, and in the absence of any vehicle. Catalysts used were metallic iron and six iron compounds. Addition of hydrogen sulfide markedly increased the catalytic activities of all catalysts except iron chloride and sulfide. Ferrous sulfide may be the effective catalyst in all instances. Unlike the other catalysts, ferrous chloride may operate by a different mechanism. Considerably more liquefaction of coal was obtained with a coal-hydrogenation vehicle oil than with the paraffinic vehicle or in the absence of a vehicle. Work is also in progress on iron catalysts impregnated on either coal or an inert support, such as graphite.

Preliminary results on the iron-catalyzed hydrogenation of asphaltene indicate that the iron chlorides have a unique activity among iron catalysts in this reaction. The mechanism may involve homogeneous catalysis by dissolved ferrous chloride.

A study has been made of the hydrogenolysis of phenyl ether at 400° C. and 500 atmospheres of hydrogen with ferrous sulfide, metallic iron, and ferrous chloride catalysts. Ferrous chloride was inactive for the hydrogenolysis of phenyl ether. Further studies of this nature are being made with benzyl and alkyl ethers.

The reaction of coal with ammonia was investigated at 400° to 500° C. and pressures up to more than 400 atmospheres. Nitrogen-containing organic compounds were formed, but ammonia did not cause any greater liquefaction of coal than was obtained by pyrolysis in an inert atmosphere.

Autoclave data indicated that high conversions of coal to a distillable oil with low asphaltene yields could be obtained in the pilot plant at about 460° C. and 2,500 p.s.i. in the presence of acid-promoted molybdenum catalyst, although the throughput would probably be lower than in commercial high-pressure processes.

Pilot Plant

Low-Pressure Hydrogenation

The advent of the industrial gas turbine and jet aircraft may cause a shift of demand from gasoline to higher boiling distillates, which may also be used eventually for domestic and industrial heating. Production of such distillates by hydrogenation in a single-stage, liquid-phase hydrogenation at low pressures would be more favorable economically than conventional hydrogenation as smaller quantities of hydrogen and simpler plants would be required.

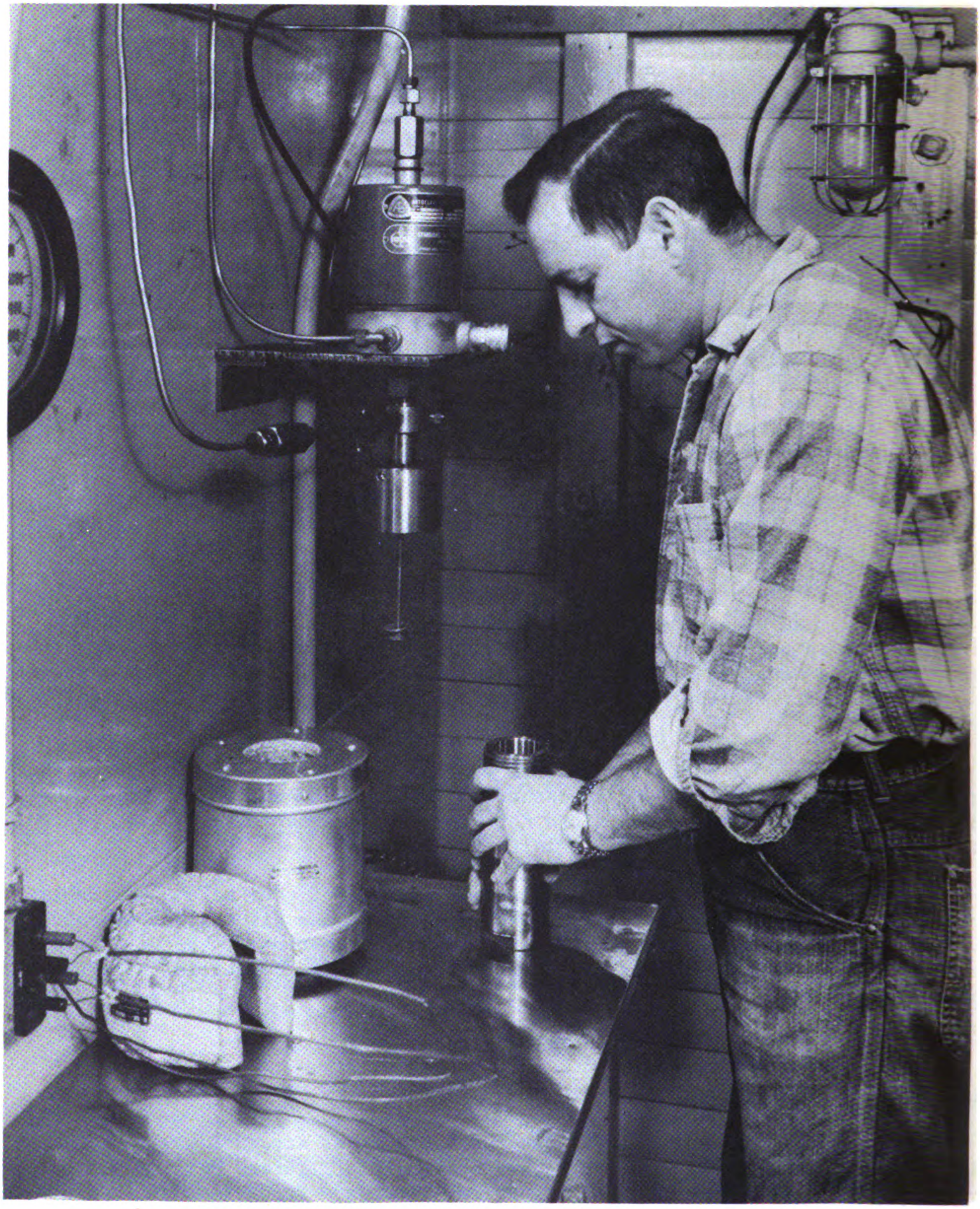

FIGURE 18. - Components of Autoclave for High-Pressure Reactions.

Because it was anticipated that the throughput at the lower pressure would be decreased, the existing high-pressure pilot plant was modified by installation of additional reactors (fig. 19). A program was developed to find the throughput and catalyst concentrations required for satisfactory conversion of coal and primary coal-hydrogenation products at low pressures to appreciable yields of distillable oils with minimum production of gaseous hydrocarbons.

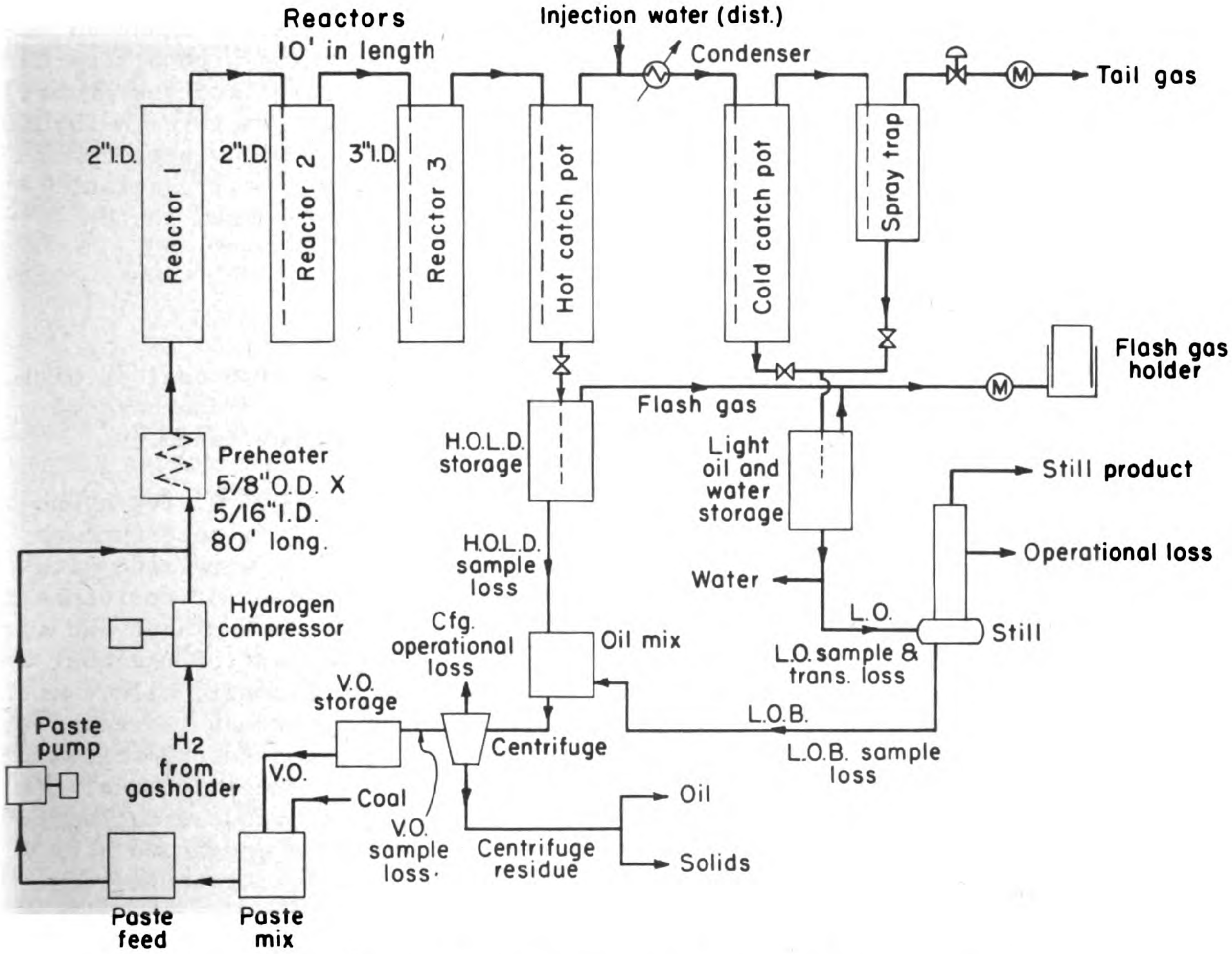

FIGURE 19. - Pilot Plant for Hydrogenating Coal at Relatively Low Pressures.

On completion of exploratory tests a study was made to determine the effect of throughput on product distribution at 2,000 p.s.i., 465° C., with 0.5 percent molybdenum (as ammonium molybdate) on coal at throughputs of 17.3, 21.5, and 24.1 pounds of coal per cubic foot of reaction space per hour. Operations with coal paste containing 40 to 45 percent coal were successful, and no difficulty was experienced in handling the high molecular products that were recycled with the fresh coal feed. The overall conversion of coal to liquid and gaseous products was 95 percent, regardless of throughput. The yield of liquid products increased only slightly as throughput increased, and the yield of gaseous hydrocarbons decreased. The physical character of the recycle-oil stream was very sensitive to throughput. Virtually all of the ash was removed from the recycle stream with no difficulty, and adequate quantities of unreacted organic matter were removed to maintain a reasonably low steady-state burden of solids.

Efforts also were made to lower the concentrations of catalyst, because the use of 0.5 percent molybdenum on coal would be uneconomical without an efficient low-cost technique for recovering catalyst. Operation with 0.1 percent molybdenum was possible by reducing throughput to 14 pounds of coal per cubic foot of reaction space per hour, about one-half of the rate possible in 10,000-p.s.i. operations. Reduction in throughput resulted in a lower viscosity of the recycle stream, so that the solids could be removed by centrifugation. Results were similar to those obtained with 0.5 percent molybdenum at the higher throughput.

High-Temperature Hydrogenation

The production of high-B.t.u. gas by reacting hydrogen with coal at high temperatures has been investigated in several laboratories over the past 15 years. A review of these hydrocarbonization processes was published.73/

The possibility of using very short reaction times at 800° C. for hydrogenating coal to low-boiling oils and gaseous hydrocarbons in a once-through operation was investigated in a semicontinuous unit. Studies were made with Rock Springs, Wyo., coal (bituminous C) to determine the effect of residence time at 800° C. and 6,000 p.s.i.g.74/ At a residence time of 1 minute and with 1 percent molybdenum (as ammonium molybdate) on coal 48 percent of the coal was converted to gaseous hydrocarbons and about 10 percent to aromatic oils. With a retention time of 15 minutes, the yield of gaseous hydrocarbons increased to 69 percent, but the yield of oil remained constant. In all cases 90 volume-percent of the hydrocarbon gases was methane and about 10 percent ethane. In the absence of a catalyst the yields of gaseous and liquid products decreased substantially at a residence time of 1 minute, but they were unchanged at a retention time of 15 minutes. A small continuous unit for hydrogenating dry coal is being constructed.

Characterization of Hydrogenation Products

Mass-spectrometric analysis of liquid coal-hydrogenation products has been extended by using a low-ionizing voltage. By this method, the types of compounds present and the molecular weights of the hydrocarbon homologs can be determined simply by inspecting the mass-spectrometer chart. These data combined with a quantitative type analysis provide a rapid method for investigating

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73/ Newman, L. L., and Pipilen, A. P., Hydro-Carbonization of Coal for High B.t.u. Gas. Part I: Gas Age, May 16, 1957, pp. 16-21; Part II. Gas Age, May 30, 1957, pp. 18-20, 44.

74/ Hiteshue, R. W., Anderson, R. B., and Schlesinger, M. D., Hydrogenating Coal at 800° C.: Ind. Eng. Chem., vol. 49, No. 12, December 1957, pp. 2008-2010.

complex, high-boiling, coal-hydrogenation fractions. The low-ionization-voltage method has been shown to be valuable in characterization of the phenolic constituents of coal-hydrogenation products and of low-temperature tar.

For characterization of synthetic fuels, new techniques of gas chromatography have been developed. With suitable liquid substrates, complicated mixtures can be separated. Other developments include the use of molecular sieves to separate branched from straight-chain isomers, the use of silver salts and ethylene glycol to separate olefins, and the preparation of trimethylsilyl ethers to aid in identifying alcohols. Chromatographic apparatus that can be used for materials with boiling points up to 450° C. has been developed for operation at 300° C. The use of rhenium as an electron emitter for mass-spectrometer ion sources has resulted in greater stability and more efficient use of the instrument for analyzing samples containing high percentages of aromatic compounds, such as coal-hydrogenation products.

Fischer-Tropsch and Related Syntheses

An alternative to the coal-hydrogenation method for producing synthetic liquid fuels involves intermediate gasification of coal to a synthesis gas containing carbon monoxide and hydrogen, with subsequent conversion to liquid products by catalytic reactions.

Chemistry of Metal Carbonyls

Investigations in carbonyl chemistry have elucidated the structure of a number of unusual compounds, many of which are similar to intermediates that have been postulated in the Fischer-Tropsch synthesis. A major aim of this work during the past year has therefore been to determine the manner in which new carbon-to-carbon bonds are formed in reactions catalyzed by complexes of iron and cobalt. A study of the cobalt carbonyl and iron carbonyl-catalyzed addition of $CO+H_2$ to olefins has led to the conclusion that these reactions involve the formation of a complex between the olefin, binuclear metal carbonyls, and hydrogen.[75 76 77] This mechanism implies that substrate CO and H_2 are brought together on two metal atoms in a "surface complex" and that chain lengthening involves the three molecules gathered together in this fashion.

From a study of the reaction between dicobalt octacarbonyl with an acetylene the first direct proof was obtained that the addition of carbon monoxide

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to a substrate can proceed via a complex.[78 79] Data were also obtained supporting the inference that complexes involving more than two metal atoms lead to the high-molecular-weight products obtained in some types of Fischer-Tropsch reactions. Furthermore, the first clear evidence that an aromatic compound can be formed by the catalytic action of a metal carbonyl was found.

Catalyst Studies

In studies of catalysts in the Fischer-Tropsch synthesis it has been shown, by measurements of surface area and pore volume, that the geometry of the catalysts changes during pretreatment and synthesis. After synthesis, iron catalysts have low surface areas and pore volumes, resulting partly from formation of iron oxide during the synthesis and partly from deposition of wax in the pores of the catalyst. That the mechanism of the synthesis involves absorption of gas in liquid hydrocarbons in catalyst pores and diffusion of this gas to the catalyst surface is supported by values of diffusivity determined from analysis of data obtained in studying the activation energies as a function of particle size of the catalyst. The physical and chemical nature of iron catalysts is considerably altered by wax and carbon deposition and by formation of carbides and magnetite.[80 81]

The Bureau has investigated the changes in the surface areas and pore volumes of iron catalysts resulting from varying the sequence of oxidation, reduction, carbiding, and nitriding and from subsequent use in the synthesis itself. Nitrided catalysts acquire appreciable amounts of carbon and become iron carbonitrides after a short period of operation.[82]

Various forms of catalyst have been studied for the hot-gas-recycle process, which requires a catalyst with a high surface-volume ratio and a minimal resistance to the flow of gases. Pretreated sheet-steel catalysts have been developed that have a low but satisfactory activity, although disintegration of the catalyst surface during a test proved to be troublesome. Some progress has been made in preparing and stabilizing the surface. The activity of such catalysts is confined to a thin layer, possibly only a few hundredths of a millimeter thick, so that most of the material is making little or no contribution to the overall catalytic activity.

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Sulfur Poisoning

Studies have been made of the poisoning of an iron catalyst by adding sulfur compounds to synthesis gas - gas-phase poisoning - and by pretreating the catalyst in a solution of the compound prior to its use - liquid-phase poisoning. The compounds studied have included hydrogen sulfide, sulfur dioxide, carbon oxysulfide, and carbon disulfide. The poison absorbed from the gas phase was 20 or more times as deleterious as that from the liquid phase; 0.1 mg. or less reduced the activity 50 percent.

Pilot Plant

In the hot-gas-recycle method of Fischer-Tropsch synthesis the large exothermic heat of the reaction is absorbed by large volumes of gas circulating through the reactor in direct contact with the catalyst. With this reactor, no heat-exchange surface is necessary in the catalyst bed, simplifying construction and decreasing the cost of reactors (fig. 20). The sensible heat of the recycle

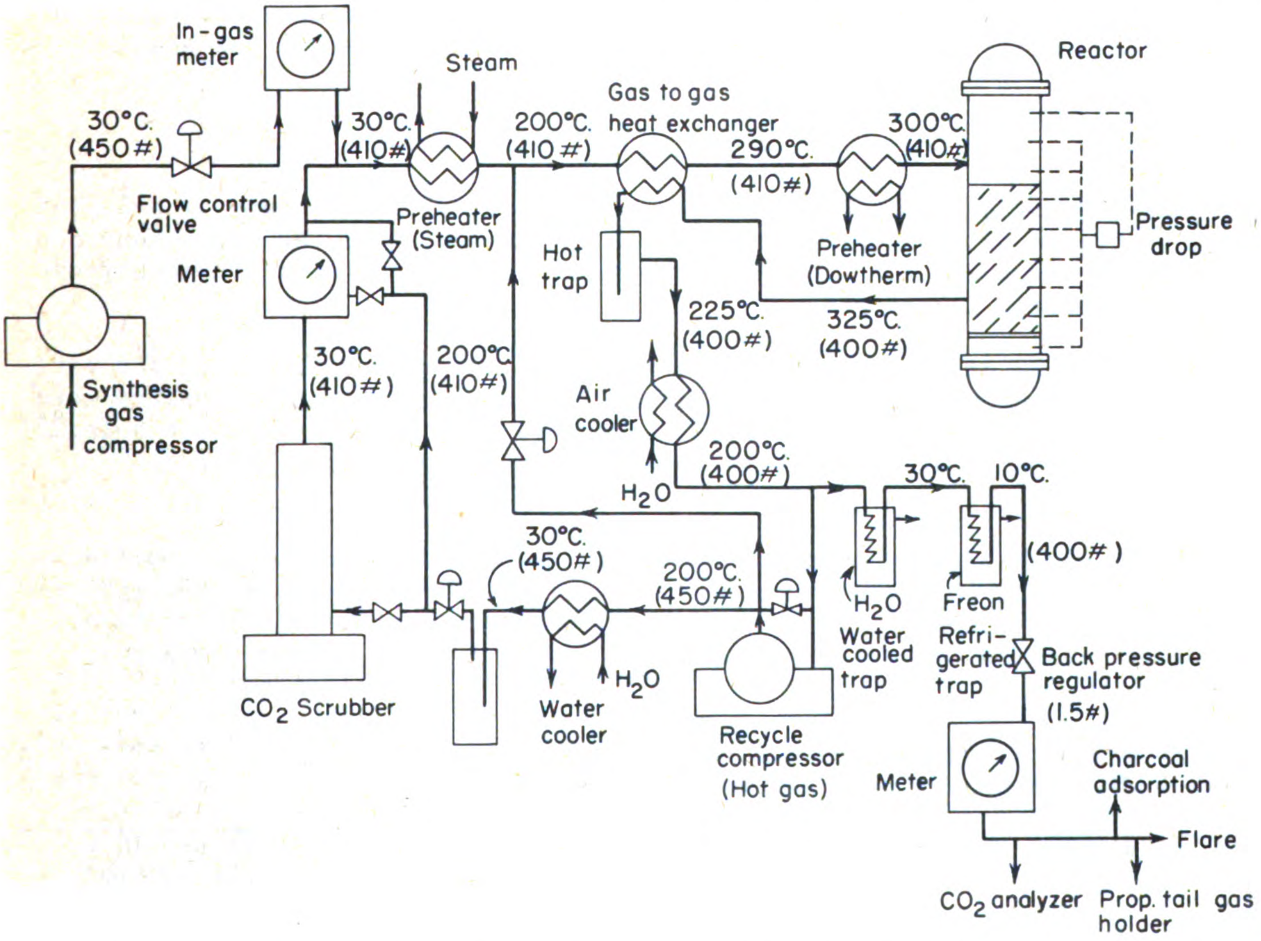

FIGURE 20. - Pilot Plant for Fischer-Tropsch Synthesis by Hot-Gas-Recycle Process.

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gas is removed by generating high-pressure steam in an external waste-heat boiler. In addition, this system can be operated readily at high temperatures, where high productivity of gasoline can be obtained. Special fixed catalyst beds are needed to maintain a low pressure drop, as the cost of recycling the gas would be excessively high with beds of granular catalyst.

The hot-gas-recycle process was operated for 85 days during the year with a few mechanical difficulties. For more than half of this time conversion of 90 to 93 percent of a feed gas containing 1.3 H_2+1 CO was achieved at an hourly space velocity of 1,000. The catalyst was prepared from lathe turnings of 1018 carbon steel; its activity remained constant through most of the test. The pressure drop across the bed was low, increasing slowly from 1 to 1-1/2 p.s.i. per foot of bed. Temperature control was excellent and there was no evidence of overheating the catalyst. During the operation, 190 pounds of hydrocarbon product (C_3+) was made per pound of iron catalyst. The experiment ended after 2,000 hours, when a leak developed in the cylinder head of the recycle compressor. Although the experiment was made at high temperatures (300° to 320° C.) and high space velocities to increase gasoline production and throughput, this type of reactor can obviously be operated at lower temperatures. During the year, a summary of the oil-circulation Fischer-Tropsch process has been published.[83]

Characterization of Synthesis Products

Gas-chromatography techniques have been applied to the study of selected light fractions of Fischer-Tropsch products that were prepared under many conditions. The results agree with the mechanism of chain growth previously developed in Bureau laboratories.

Correlation of mass spectra with molecular structures continued for various types of compounds present in Fischer-Tropsch product. Mass spectra were published of 26 aliphatic trimethylsilyl ethers used in analyzing alcohols.[84] Correlation studies have also been completed on the mass spectra of 31 aliphatic esters.

Researchers have investigated a liquid Fischer-Tropsch product boiling above 100° C. both by room-temperature and high-temperature mass spectrometers. Complete carbon-number distributions have been obtained for straight-chain paraffins, olefins, ketones, and alcohols having 2 to 22 carbon atoms per molecule.

By means of molecular-sieve techniques the ratio of branched to straight-chain hydrocarbons has been determined in a fraction consisting primarily of

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C_{13} and C_{14} hydrocarbons. Results show the sample to be 1/3 by volume normal hydrocarbons, which fact agrees with values predicted by the Fischer-Tropsch chain-growth scheme.

Reviews of Literature

In connection with the Bureau's studies of the Fischer-Tropsch and related processes a book review and four contributions to reference works were published.85 86 87 88 89/

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